Effects of rural domestic sewage reclaimed irrigation and regulation on heavy metals, PPCPs, water and nitrogen utilization, and microbial diversity in paddy field

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Abstract: Rural domestic reclaimed water (RDRW) is rural domestic sewage that being safely treated, the irrigation and reuse of RDRW are an effective way to alleviate the contradiction between supply and demand of water resources in South China. In this study, four kinds of irrigation water sources (primary and secondary treated water R1 and R2, purified water R3 and river water CK) and three kinds of water level regulations (low, medium, and high field water level control of W1, W2 and W3) were set to study the impact of RDRW on soil and crop safety, water and nitrogen utilization and biodiversity for establishing the regulation mechanism of RDRW irrigation with field experiment, and monitoring was carried out in RDRW irrigation demonstration area to assess the effectiveness of RDRW. The results showed that, under RDRW irrigation, the contents of Cd and Pb increased slightly, while the contents of Cr, Cu and Zn decreased in paddy soil. The heavy metals content decreased along the direction of stem, leaf and grain in rice plants, but did not increase significantly in rice grains. With the increase of field water level, pharmaceutical and personal care products (PPCPs) content in 60-80 cm soil layer was accumulated, and the PPCPs content in rice husks was higher than that in grains, but it was at a very low level. Compared to CK, RDRW irrigation can effectively increase rice yield, rainwater use efficiency (RUE) and nitrogen use efficiency (NUE) by 5.4%-7.6%, 6.7%-9.4% and 21.7%-24.2%, respectively, and the species diversity, community diversity and richness in rice fields were improved. Additionally, water level regulation of W3 with R2 water resource irrigation was conducive to the exertion of comprehensive benefits. The monitoring of demonstration area showed that the consumption of fresh water was reduced by 530 mm, yield was increased by 9.6%, and the soil and crop were both safety. Short-term irrigation of RDRW did not cause soil and crops pollution, however, it is still necessary to track and monitor the effect of the system on soil, crop, and underground water with long-term reclaimed water irrigation.

Keywords: reclaimed water irrigation, heavy metals, pharmaceutical and personal care products, water and nitrogen use efficiency, soil microbial diversity, technique for order preference by similarity to an ideal solution
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1 Introduction

With the large consumption of water in industry and cities, the gap of agricultural irrigation water reaches 60 billion m^3 every year in China, and it is increasing year by year. The contradiction of water shortage has become increasingly prominent. With the increasing discharge of rural domestic sewage, the problem of agricultural non-point source pollution is serious. Rural domestic sewage including feces and their washing water, bathing sewage and kitchen sewage, has the characteristics as follows, small difference in water quality, dispersed water volume, high content of nitrogen (N) and phosphorus (P), good biodegradability and generally free of toxic substances such as heavy metals, compared with urban sewage. Rural domestic reclaimed water (RDRW) is rural domestic sewage that has been safely treated, which can be used as the fertilizer resource and is easier to ensure the safety of irrigation reuse[1,2]. As a food crop with high water consumption, rice is an important carrier of regional ecological environment, and it can maximize the consumption and utilization of domestic sewage[3-5]. The core of irrigation regulation of RDRW is to improve the water and nitrogen utilization efficiency, and promote rice yield by changing soil environmental quality and habitat characteristics[6].

Scholars in China and abroad have done a lot of researches on the effects of reclaimed water irrigation process on physical and chemical properties of soil and crop yield. Liu et al.[7] and Khamisi et al.[8] found that winter wheat and maize had obvious yield increase effect with reclaimed water irrigation. Alkhamisi et al.[9]

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found that the cotton yield under reclaimed water irrigation was equivalent to that of clean water irrigation with equal nitrogen input, which was conducive to nitrogen transformation and crop absorption and utilization. A large number of studies have shown that reclaimed water irrigation can effectively improve nitrogen availability and increase the activities of enzymes-related and microorganisms-related transformation of N and P in soil. Chen et al.[10] found that reclaimed water irrigation could increase soil nutrient content and improved soil's microbial activity and diversity. Cheng et al.[11] showed that about 34% of the nitrogen entering the system with reclaimed water irrigation could be absorbed and utilized by crops, and 62% could be removed or adjusted the amount of nitrogen in soil nitrogen pool by denitrification.

With the complex behavior process, heavy metal pollution is a hot issue in the research and application of reclaimed water reuse, in which all kinds of physical, chemical and biological reactions in soil, as well as the participation of crops are involved. The common heavy metals in rural domestic sewage include Cu, Cr, Pb, Cd and Zn, and their concentration is usually below 5 μg/L, among which Cr and Cd have a higher ecological risk[12,13]. With the acceleration of urbanization and the improvement of living standards in urban and rural areas, the pollution of emerging pollutants of pharmaceutical and personal care products (PPCPs) increases rapidly. Agricultural and animal husbandry, household and hospital wastewater, being the main sources of PPCPs, could directly or indirectly pollute the soil through the discharge of sewage disposal, and finally enter the surface or underground water body. The PPCPs contents, such as acetaminophen, ibuprofen and diclofenac, are higher, up to more than 50 μg/L, the concentration range of caffeine is 0.5-200 μg/L, and the concentration of pesticides and surfactants are 20-100 μg/L[14,15]. Different irrigation methods and water quality characteristics have different effects on the migration and accumulation of heavy metals and PPCPs in soil and crop system. Therefore, analyzing the migration and transformation of heavy metals and PPCPs and evaluating the comprehensive ecological environment impact under farmland irrigation regulations are key to ensure the safe reuse of sewage.

At present, the utilization of RDRW is still at a relatively initial stage. The research on the regulation mechanism of reclaimed water irrigation based on the safety of soil and crop system, efficient utilization of water and nitrogen, soil microbial diversity and high yield of crops is still insufficient. The objectives of this study mainly included, (1) to analyse migration and accumulation of heavy metals and PPCPs, reclaimed water use efficiency and nitrogen utilization rate under different grades of treated sewage water and water level regulations, (2) to establish an efficient and safe reclaimed water irrigation regulation mechanism, (3) and to evaluate the comprehensive benefits in the demonstration area.

The results have important theoretical and practical significance for formulating a scientific and reasonable safe and efficient operation and management mode and realizing the healthy and sustainable development of reclaimed water irrigation.

2 Materials and methods

2.1 Experimental field

This study was carried out in Yongkang rural domestic sewage recycling base (28°48'N, 120°10'E) from May to October in 2020 and 2021. The study area belongs to the landform of low mountains and hills, where is of the subtropical monsoon climate, with average annual precipitation of 1787 mm, maximum annual precipitation of 2389 mm and minimum annual precipitation of 1120 mm. In the local area, the annual average evaporation is 930 mm, the annual average temperature is 17.5°C, the annual maximum and minimum temperature is 39.9°C and –14.5°C, the annual average sunshine is 1909 h, and the frost free period is 245 d. The study area included a field experimental area and a demonstration area. Field experimental area is composed of 36 standard experimental plots (size 20 m×5 m per plot), thus the demonstration area is about 26.67 hm^2, which is mainly planted with rice and cash crops. For the location and layout of the study area, see Figure 1, and for the environmental quality indicators of different soil layers, see Table 1.

pH, EC, WSS, TN, TP, NH_4^+-N, and NO_3^--N represented soil acidity and alkalinity, electrical conductivity, water soluble salt, total nitrogen, total phosphorus, organic matter, ammonium-nitrogen and nitrate-nitrogen, respectively.

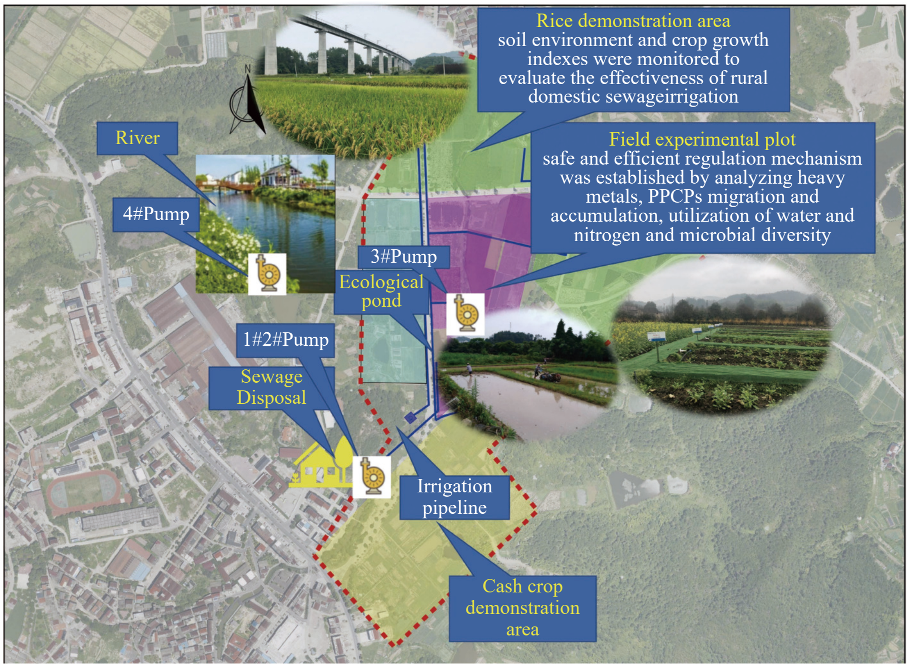


Figure 1 Location and layout of the study area

Table 1 Soil environmental quality indicators in study area

Soil depth/mm	pH	EC/ $mS\cdot m^{-1}$	WSS/ $g\cdot kg^{-1}$	TN/%	TP/%	OM/ $g\cdot kg^{-1}$	NH_4^+-N/ $mg\cdot kg^{-1}$	NO_3^--N/ $mg\cdot kg^{-1}$	Cr/ $mg\cdot kg^{-1}$	Cu/ $mg\cdot kg^{-1}$	Cd/ $mg\cdot kg^{-1}$	Pb/ $mg\cdot kg^{-1}$	Zn/ $mg\cdot kg^{-1}$
0-20	5.56	2.6	0.44	0.12	0.069	17.7	8.24	2.84	20.92	8.67	0.06	36.67	74.08
20-40	5.88	2.9	0.27	0.09	0.032	14.8	5.75	2.69	22.67	8.58	0.05	34.08	72.42
40-60	6.15	2.8	0.26	0.07	0.027	12.9	4.71	2.50	22.42	8.58	0.05	35.58	78.33
60-80	6.61	2.8	0.26	0.04	0.03	8.0	3.17	2.16	23.00	6.92	0.03	33.33	71.25

In the study area, a domestic sewage disposal with design scale of 400 m^3/d has been built, which was studied as the rural domestic sewage source in this study. The treatment process adopted the secondary treatment process. Primary treatment was conventional process, namely physical precipitation method, mainly to remove suspended solid substances in sewage, and the secondary treatment process adopted Anaerobic+Anoxic+Oxic (A^2/O), which had the function of simultaneous N and P removal, and the effluent quality met the class I A standard in the discharge standard of pollutants for urban sewage treatment plants (GB18918-2002)[16]. An ecological pond with a sewage storage capacity of 3000 m^3 was built to store and purify rural domestic sewage. The demonstration area transmitted reclaimed water to the paddy field by improving the sewage treatment terminal.

2.2 Experimental design

Four kinds of irrigation water sources were set in the field experimental area, namely primary treated water (R1) and secondary treated water (R2) from the sewage disposal, purified secondary treated water by ecological pond (R3), and river water (CK). In 2020, the ecological pond has not been built yet, and the water source is temporarily replaced by river water. Therefore, there are three kinds of irrigation water sources (R1, R2 and CK) in 2020, while in 2021, the ecological pond water is supplemented for irrigation, and there are four kinds of irrigation water sources. The statistics of irrigation water quality indicators during the experimental period are listed in Table 2, and all indicators met the standards for irrigation water quality (GB5084-2021)[17]. Farmland water level refers to the water depth maintained by farmland after

Table 2 Description and statistics of water quality indicators of different irrigation water sources (mg/L)

Water sources	Indicator	Maximum value	Minimum value	Standard deviation	Mean value	Kurtosis	Skewness
R1	COD	84	15	26.794	29.5	5.855	2.410
	LAS	0.88	0.06	0.315	0.25	5.199	2.247
	NH_4^+-N	11.9	8.25	1.645	9.647	–1.782	0.916
	NO_3^--N	0.061	0.016	0.019	0.034	–1.452	0.642
	Cr	1.51	0.34	0.59	1.00	–4.41	–0.28
	Cu	19.92	2.39	8.21	7.67	3.80	1.93
	Cd	0.06	0.00	0.02	0.02	2.03	1.18
	Pb	0.43	0.22	0.09	0.30	1.98	1.40
	Zn	35.23	22.43	5.73	30.12	–0.06	–1.00
R2	COD	59	10	16.783	24.1	0.719	1.291
	LAS	0.16	0	0.058	0.048	–0.425	0.827
	NH_4^+-N	11.9	3.52	2.837	7.712	–0.946	–0.174
	NO_3^--N	6.25	0.01	2.455	1.364	1.238	1.687
	Cr	2.48	0.23	0.97	1.10	2.62	1.42
	Cu	7.53	1.00	2.82	3.46	2.85	1.53
	Cd	0.02	0.00	0.01	0.01	1.58	–0.85
	Pb	1.53	0.17	0.59	0.81	–0.78	0.36
	Zn	40.71	12.65	13.11	22.29	1.24	1.36
R3	COD	62	11	12.735	24.15	0.710	1.553
	LAS	0.32	0	0.132	0.042	–1.215	0.826
	NH_4^+-N	5.45	2.34	0.634	4.415	0.478	0.473
	NO_3^--N	3.16	0.345	0.928	0.823	1.382	1.275
	Cr	1.33	0.22	0.46	0.73	1.22	0.62
	Cu	3.44	0.88	1.09	2.00	0.43	0.76
	Cd	0.10	0.00	0.05	0.03	3.81	1.94
	Pb	1.11	0.11	0.44	0.56	–0.86	0.59
	Zn	20.33	5.68	6.51	11.11	1.79	1.39
CK	COD	56	7	15.712	23.45	0.710	1.251
	LAS	0.1	0	0.041	0.035	–1.875	0.418
	NH_4^+-N	1.49	0.116	0.394	0.711	0.143	0.393
	NO_3^--N	2.56	0.624	0.578	1.048	4.680	2.078
	Cr	0.94	0.15	0.32	0.57	1.54	–0.43
	Cu	2.65	0.73	0.86	1.59	–1.93	0.47
	Cd	0.01	0.00	0.01	0.01	2.10	–1.20
	Pb	0.24	0.14	0.05	0.20	–1.50	–0.72
	Zn	18.23	12.98	2.69	15.85	–5.04	–0.16

Note: COD, LAS, NH_4^+-N, and NO_3^--N represented chemical oxygen demand, linear alkylbenzene sulfonates, ammonium-nitrogen and nitrate-nitrogen, respectively.

rainfall or irrigation and the buried depth of farmland groundwater level when there is no water layer. In this study, the farmland water level was used as the control indicator of irrigation and drainage in paddy field, and three kinds of water level regulation were set, with three repetitions for each treatment. The water level regulation in paddy field was strictly controlled in each growth period of rice. When the water level dropped to the lower limit, the reclaimed water will be replenished. In case of rainstorm exceeding the upper limit of rain storage, the rainwater will be drained. The field water level control standards are listed in Table 3. There were twice fertilization for all treatments during the growth, i.e., the basal fertilizer of 200 kg/hm² compound fertilizer (N:P:K=18:8:15) and 100 kg/hm² urea (with nitrogen content of 47%) on Jun 25th and July 6th, and the dressing fertilizer of 250 kg/hm² compound fertilizer on July 12th and July 18th in 2020 and 2021, respectively. For the experimental layout, see Figure 2. The irrigation water source and water level regulation in the demonstration area were controlled by the optimal control scheme in the field experimental plot, and the fertilization method was consistent with that in the field experimental plot.

Table 3 Standard of water level control in paddy field (mm)

Water control	Upper and lower limit	TG	ET	LT	JB	HF	MK
W1	Upper limit of sewage	0	3-5 d exposing field	1-2 d exposing field	1-2 d exposing field	1-2 d exposing field	3-5 d exposing field
	Lower limit of sewage	30	30	exposing field	40	40	30
	Upper limit of storage	50	70		80	80	60
W2	Upper limit of sewage	0	10	10	10	10	10
	Lower limit of sewage	30	50	exposing field	50	50	50
	Upper limit of storage	50	70		100	100	100
W3	Upper limit of sewage	0	40	40	40	40	10
	Lower limit of sewage	30	60	exposing field	60	60	60
	Upper limit of storage	50	100		150	150	100

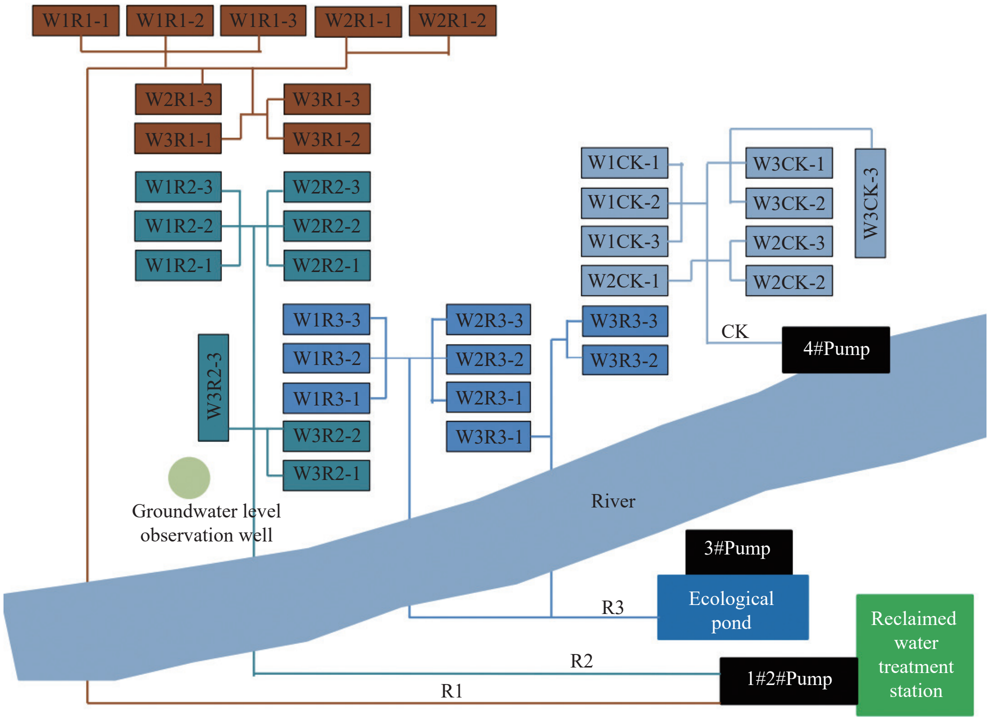


Figure 2 Experimental layout of paddy field.

Standard deviation was the arithmetic square root of variance, reflecting the dispersion of a data set, mean value was the average value of a data set, kurtosis was a statistic that describes the gradient of all value distribution patterns, and skewness was a statistic that describes the distribution form of data, reflecting the symmetry of a data set.

TG, ET, LT, JB, HF and MK represented turning green, early tillering, later tillering, jointing and booting, heading and flowering, milky of rice growth stages, respectively. 1-2 d and 3-5 d exposing field indicated 1 mm and 2 mm cracks in the paddy field after exposing field.

2.3 Indicators and measurements

Before the experiment and after the rice harvest of each year, samples were taken from different soil layers (0-20 cm, 20-40 cm, 40-60 cm and 60-80 cm) and rice plants (stems, leaves, grains, rice husks) in each experimental plot and demonstration area. Soil samples were collected by using the five-point method, and the content of Pb, Cr, Cu and Zn were determined by the flame atomic absorption spectrophotometry, Cd content was determined by graphite furnace atomic absorption spectrophotometry, PPCPs were determined by combining chromatography-mass and mass spectrometry method, and microbial diversity indexes were sequenced by MISEQ sequencing platform (Illumina, US). For plant samples, they were sampled once per growth period by using the five-point method. The content of Cu and Zn were determined by flame atomic absorption spectrophotometry, and the content of Pb, Cr and Cd were determined by inductively coupled plasma mass spectrophotometry, and the determination method of PPCPs content was the same as that of soil samples. The actual yield of different treatments was measured after rice harvest.

In the field experimental plot, water consumption (WCT) was measured by the change of water level in field surface according to the measuring needle on 2 consecutive days when there is a water layer, while it was measured by the change of soil water content according to the soil moisture analyzer at 8:00 am every day when there is no water layer. Water leakage was measured by the leakage meter on 2 consecutive days, and then the leakage amount was calculated according to the reading difference by the measuring needle before and after that. The crop water consumption was the difference between WCT and leakage. In the demonstration area, irrigation water amount (IWA) was read by the electromagnetic flowmeter (ZNK-6, China) installed in each lift pump station, and the leakage rate of irrigation pipeline was taken as 5%.

Water use efficiency (WUE) was defined as yield per unit of water consumption and calculated from the ratio of yield to water consumption. WUE could be divided into $\mathrm{WUE_I}$ and $\mathrm{WUE_{ET}}$ when the water consumption represented IWA, and crop water consumption, respectively. RUE (rainwater use efficiency) was the ratio of effective rainfall to total rainfall. NUE (Nitrogen use efficiency of reclaimed water) was the ratio of nitrogen brought in by reclaimed water absorbed by crops and soil residues to the total nitrogen brought in by reclaimed water.

2.4 Statistical analysis

The potential ecological risk index (RI) was used to analyze the potential ecological risk of heavy metals in farmland soil irrigated by different water sources in the area of study. The calculation formula is as followed.

$$RI=\sum E_i=\sum T_i\cdot P_i$$

where, RI is the comprehensive potential ecological risk index of various heavy metals in soil, E_i is the single factor ecological risk coefficient of each evaluation index in soil samples, Pi is the single factor pollution index in the soil evaluation index, which is the ratio of the measured value of the single pollution factor to the corresponding background value, and Ti is the toxicity coefficient of heavy metals in soil.

In this paper, a multi-objective decision model of technique for order preference by wimilarity to an ideal solution (TOPSIS) was established to optimize the regulation schemes of different irrigation water sources and water level regulations[18]. The basic idea is to determine the ideal solution and negative ideal solution of the decision by constructing the weighted standardized decision-making matrix of the evaluation index value, and then calculate the Euclidean distance between the evaluated scheme and the ideal solution and the negative ideal solution, so as to determine the relative closeness between the evaluated scheme and the ideal scheme, and finally select the scheme closest to the ideal solution as the optimal decision. The modeling and solving steps are as follows:

Step 1: evaluated samples and indexes are assumed as n and m, so this evaluation system can be indicated by the initial evaluation matrix $[Y]=(y_{ij})_{n\times m}$, and then make a normalized treatment to get a normalized matrix $[R]=(r_{ij})_{n\times m}$, among them, the indicators for the larger and better income can be calculated by the formula as $r_{ij}=(y_{ij}-\min y_{ij})/(\max y_{ij}-\min y_{ij})$, while the indicators for the smaller and better cost indicators can be calculated by the equation as $r_{ij}=(\max y_{ij}-y_{ij})/(\max y_{ij}-\min y_{ij})$.

Step 2, the entropy of the index of the number j (j=1,2,…,m) can be defined as $\mathrm{e}_j=-k\sum_{i=1}^{n}f_{ij}\ln f_{ij}$, among them, $f_{ij}=R_{ij}/\sum_{i=1}^{n}R_{ij}$, k=1/ln$n$, and the corresponding entropy can be defined as $\omega_j=(1-e_j)/(m-\sum_{j=1}^{m}e_j)$.

Step 3, a weighted standardized decision matrix is constructed as $[Z]=(z_{ij})_{n\times m}$, which is calculated by the formula as $z_{ij}=\omega_j\times r_{ij}$, $(i\in n, j\in m)$.

Step 4, ideal solution and negative ideal solution are determined as $x^+=(x_1^+,x_2^+,\ldots\ldots,x_m^+)$ and $x^-=(x_1^-,x_2^-,\ldots\ldots,x_m^-)$, respectively, among them, the larger and better income indicators are $x_j^+=\max_j z_{ij}$ and $x_j^-=\min_j z_{ij}$, while the smaller and better cost indicators are $x_j^+=\min_j z_{ij}$ and $x_j^-=\max_j z_{ij}$.

Step 5, the Euclidean distance of the ideal solution and the negative ideal solution are calculated by the formula as

$$d_i^+=\sqrt{\sum_{j=1}^{m}(z_{ij}-x_j{}^+)^2}\text{ and }d_i^-=\sqrt{\sum_{j=1}^{m}(z_{ij}-x_j^-)^2}\text{, respectively.}$$

Step 6, the relative closeness between each scheme and the ideal solution is calculated by the formula as $S_i=\dfrac{d_i^-}{(d_i^++d_i^-)}$.

Finally, S_i is arranged from large to small, and the largest value is the best.

Additionally, the data calculation and diagramming were completed by EXCEL 2013. ANOVA analysis was carried out by SPSS Statistics 19. Multi objective decision model adopts TOPSIS carried out by SPSSAU.

3 Results

3.1 Safety in soil and plant of RDRW irrigation

3.1.1 Heavy metal content in paddy soil

The contents of heavy metals in paddy soil are shown in Figure 3. After irrigation in 2020 and 2021, the contents of Cd and Pb were increased, but the contents of Cr, Cu and Zn were decreased, that may be related to the RDRW mainly coming from domestic sewage, of which the contents of Cr, Cu and Zn were relatively lower. The Cd content in the surface soil layer was significantly higher than that in the deep soil layer, which was just due to the small mobility of Cd, and it was easy to accumulate in the upper layer of the soil. With the increase of water level control and the extended of irrigation time, the Cd content in each soil layer showed an obvious increasing trend. The increase of Cd content in each soil layer was largest under R1, moderate under R2, and smallest under R3, where the Cd content under R3 was close to that under CK. Such results indicated that the ecological pond had a certain interception effect on Cd after purification. In the year of 2020, Cr content in paddy field was decreased, except for the treatments of W1R2 and W3R2, but the Cr content was generally increased compared with the background value in 2021. With the extended irrigation time, the Cr content changed significantly. The results of ANOVA are shown in Table 4. It showed that water level regulation and irrigation water source had a significant impact on Cd content in both 0-20 cm (p<0.01) and 20-40 cm soil layers (p<0.05), and irrigation water source had a significant impact on Cr content in 20-40 cm soil layer and Cu content in 40-60 cm soil layer (p<0.01). In addition, according to the calculation of the potential ecological risk index (RI) of heavy metals in the root layer (0-20 cm) in Table 5, the RI of R1, R2, R3 and CK was 129.96, 109.38, 65.34 and 69.22, respectively. It can be seen that the RI of R3 was the lowest, which was similar to that of CK. It was because the domestic sewage inflow in this area exceeded the treatment capacity, and the rural domestic sewage was discharged into river, at the same time, the purification effect of the ecological pond for R3 water source resulted in the RI of CK being slightly lower than that of R3. According to the heavy metal ecological risk rating standard, the ecological risk of each water source irrigation was mild.

3.1.2 Heavy metal content in rice plant organs

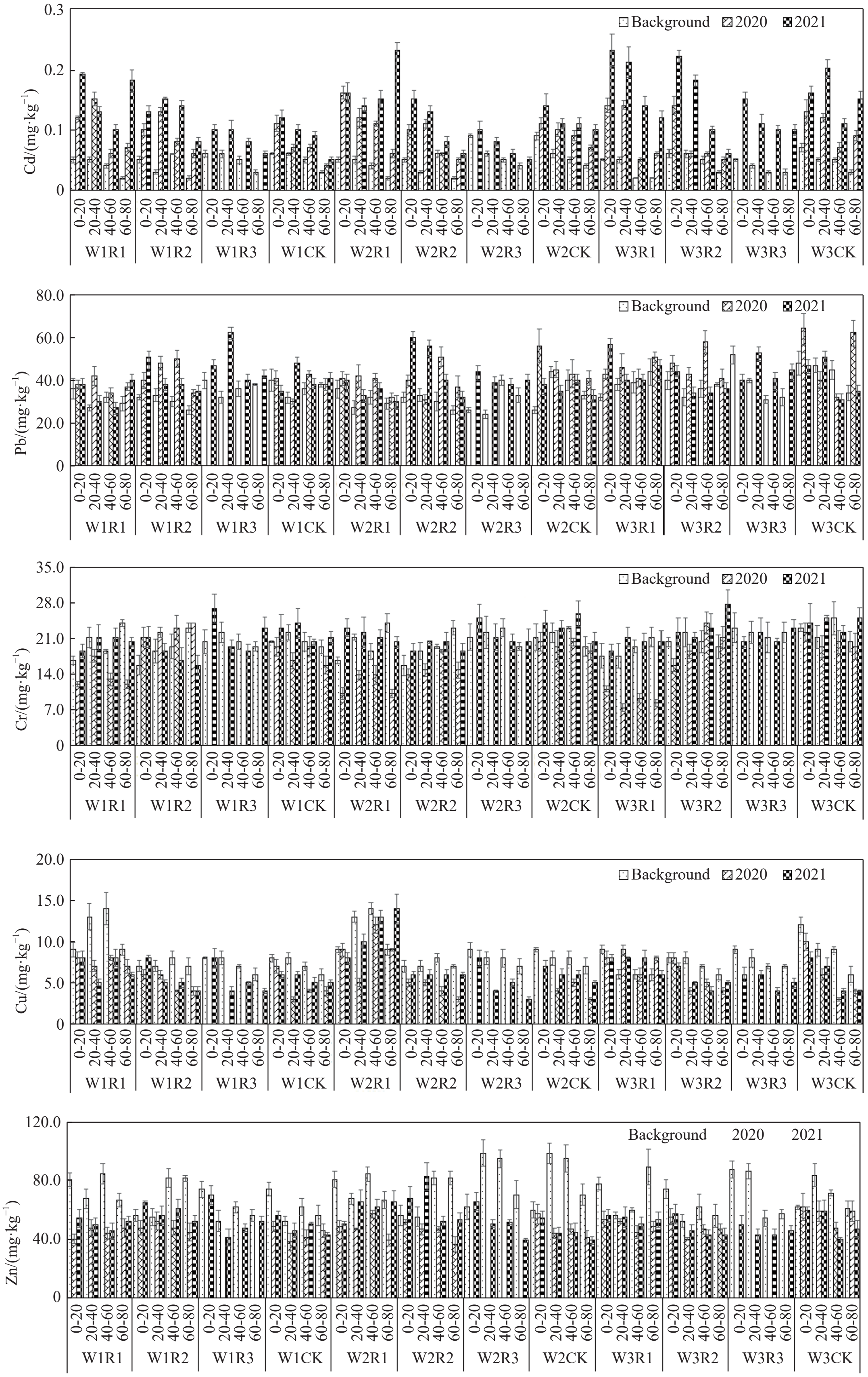


Figure 3 Content of heavy metals in paddy soil under different irrigation water sources and water level regulations.

Table 4 Two factor analysis of variance (ANOVA) of heavy metals in paddy field

Soil layer	Cd		Pb		Cr		Cu		Zn	
	W	R	W	R	W	R	W	R	W	R
0-20 cm	**	**	NS	NS	NS	NS	NS	NS	NS	NS
20-40 cm	*	*	NS	NS	NS	**	NS	NS	NS	NS
40-60 cm	NS	NS	NS	NS	NS	NS	NS	**	NS	NS
60-80 cm	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS

NS means not significant ($p>0.05$), * means $p<0.05$, and ** means $p<0.01$

Table 5 The potential ecological risk index (RI) of heavy metals in 0-20 cm layer

Water source	Cd		Pb		Cr		Cu		Zn		*RI*
	Pi	*Ei*	*Pi*	*Ei*	*Pi*	*Ei*	*Pi*	*Ei*	*Pi*	*Ei*	
R1	3.87	116.00	1.30	6.49	1.18	2.36	0.89	4.44	0.67	0.67	129.96
R2	3.13	93.75	1.49	7.45	1.20	2.39	0.95	4.77	1.02	1.02	109.38
R3	1.75	52.50	1.11	5.55	1.11	2.23	0.85	4.23	0.83	0.83	65.34
CK	1.91	57.27	1.05	5.26	1.10	2.20	0.72	3.62	0.87	0.87	69.22

Contents of heavy metals in different organs of rice plant are shown in Figure 4. It is shown that the heavy metals content decreased along the direction of stem, leaf and grain in rice plants. For rice stems, the total content of heavy metals under different irrigation water source conditions showed a decreasing trend with the irrigation water quality changes from poor (R1, R2) to excellent (R3, CK). Compared to CK, the total content of heavy metals under R1, R2 and R3 were increased by 1.52 times, 1.25 times and 1.15 times, respectively, and the total amount of heavy metals under R3 was close to that under CK. The total content of heavy metals showed a decreasing trend with the increase of water levels. Compared to W3, the total content of heavy metals under W1 and W2 were increased by 1.07 times and 1.06 times, respectively. For rice leaves, the total content of heavy metals in different irrigation water sources showed the largest under R1, moderate under CK, and smallest under R2 and R3. Compared to R3, the total content of heavy metals under R1, R2, and CK were increased by 1.07 times, 1.04 times and 1.06 times, respectively. The total content of heavy metals by different water levels showed the largest under W3, moderate under W1, and smallest under W2. Compared to W2, the total content of heavy metals under W1 and W3 were increased by 1.05 times and 1.17 times, respectively. For rice grains, the total contents of heavy metals in different irrigation water sources were consistent with that in stems. Compared to CK, the total content of heavy metals under R1, R2 and R3 were increased by 1.16 times, 1.09 times and 1.12 times, respectively, and it was basically the same though regulated by different water level regulations. The results of ANOVA are shown in Table 6. It showed that different irrigation water sources had significant effects on Zn content in both stems and leaves, and Pb content in grains ($p<0.05$), and Pb content in stems, and Cu content in both leaves and grains ($p<0.01$). Different water level regulations had a significant effect on Cd content in stems and leaves ($p<0.01$), and Cr content in leaves ($p<0.05$).

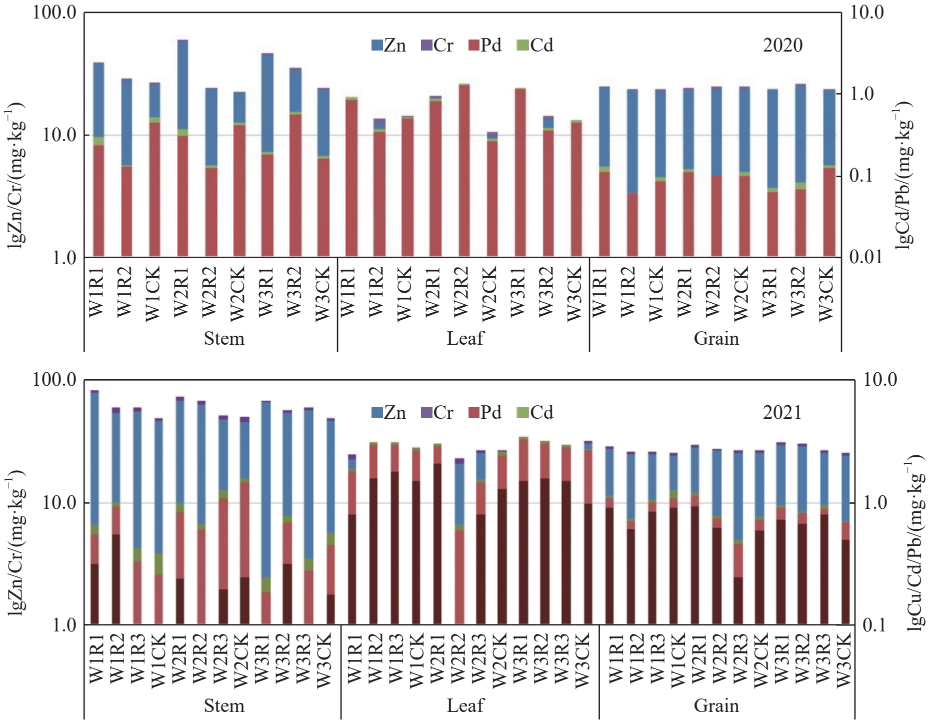


Figure 4 Content of heavy metals in rice plant organs under different irrigation water sources and water level regulation

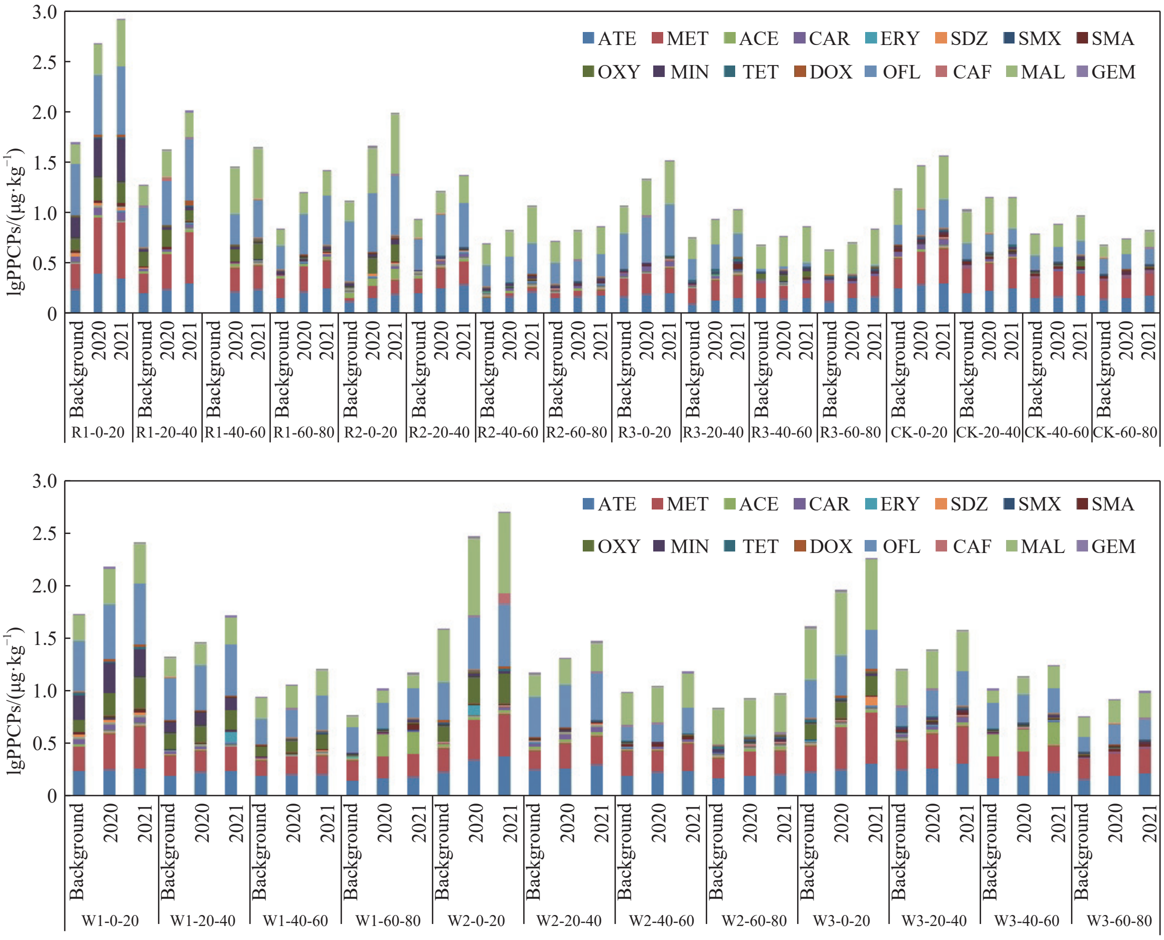
Table 6 Two factor analysis of variance (ANOVA) of heavy metals in plant organs

Organ	Zn		Pb		Cd		Cr		Cu	
	W	R	W	R	W	R	W	R	W	R
Stem	NS	*	NS	**	**	NS	NS	NS	NS	NS
Leaf	NS	*	NS	NS	**	NS	*	NS	NS	**
Grain	NS	NS	NS	*	NS	NS	NS	NS	NS	**

Note: W and R represented water level control and water sources of irrigation, respectively. NS meant not significant ($p>0.05$), * meant $p<0.05$, and ** means $p<0.01$

3.1.3 PPCPs content in paddy soil

In this experiment, 23 kinds of PPCPs were detected in paddy soil, and 16 kinds of PPCPs were considered. The changes of PPCPs content in paddy field are shown in Figure 5. There were 6 kinds of PPCPs with high content in paddy field, namely atenolol (ATE), metoprolol (MET), ofloxacin (OLF), malathion (MAL), oxytetracycline (OXY), and minocycline (MIN), with the change



ATE, MET, ACE, CAR, ERY, SLD, SLM, SLMZ, OXY, MIN, TET, DOX, OFL, CAF, MAL and GEM represent atenolol, metoprolol, acetaminophen, carbamazepine, erythromycin, sulfadiazine, sulfamethoxazole, sulfamethazine, oxytetracycline, minocycline, tetracycline, doxycycline, ofloxacin, caffeine, malathion, and gemfibrozil, respectively.

Figure 5 Changes of pharmaceutical and personal care products (PPCPs) content in paddy field by different irrigation water sources and water level regulations

range of 0.10-0.40 μg/kg, 0.02-0.55 μg/kg, 0.003-0.68 μg/kg, 0.13-0.59 μg/kg, 0.001-0.23 μg/kg, and 0.001-0.23 μg/kg, respectively. With the increase of soil depth, the PPCPs content decreased, and the interannual change of PPCPs content in each soil layer showed an increasing trend. For different irrigation water sources, the growth rate of PPCPs in paddy field was varied greatly, and showed a decreasing trend with the irrigation water quality changed from poor (R1, R2) to excellent (R3, CK), and it was similar for R3 and CK. For R1 irrigation, compared to background value (values of soil sampled before the experiment), the PPCPs content was increased by 1.58 times and 1.73 times in 0-20 cm soil layer, 1.27 times and 1.58 times in 20-40 cm soil layer, 1.50 times and 1.71 times in 40-60 cm soil layer, and 1.44 times and 1.70 times in 60-80 cm soil layer, respectively in 2020 and 2021. For R2 irrigation, compared to background value, the PPCPs content was increased by 1.48 times and 1.76 times in 0-20 cm soil layer, 1.30 times and 1.48 times in 20-40 cm soil layer, 1.20 times and 1.58 times in 40-60 cm soil layer, and 1.15 times and 1.21 times in 60-80 cm soil layer, respectively in 2020 and 2021. For R3 and CK irrigation, in 2020 and 2021, the PPCPs content was increased by 1.24 and 1.19 times, 1.41 and 1.26 times in 0-20 cm soil layer, 1.25 and 1.12 times, 1.38 and 1.13 times in 20-40 cm soil layer, 1.14 and 1.13 times, 1.18 and 1.24 times in 40-60 cm soil layer, 1.11 and 1.09 times, 1.32 and 1.21 times in 60-80 cm soil layer, respectively. For different water level regulations, the growth rate of PPCPs was increased by medium (W2) and high water level regulation (W3) in 0-20 cm soil layer, basically no obviously changed in 20-60 cm soil layer, but obviously increased by high water level regulation (W3) in 60-80 cm soil layer, indicating that the PPCPs content in 60-80 cm soil layer was accumulated with the increase of water level control in paddy field.

3.1.4 PPCPs content in rice plants

The analysis of PPCPs content in paddy field showed that different water level regulations had little impact on the PPCPs content in paddy field. Therefore, the changes of PPCPs content in rice plants were analyzed under different irrigation water sources (Figure 6). For rice grains, the high content of PPCPs mainly included atenolol (ATE), ofloxacin (OFL), malathion (MAL),

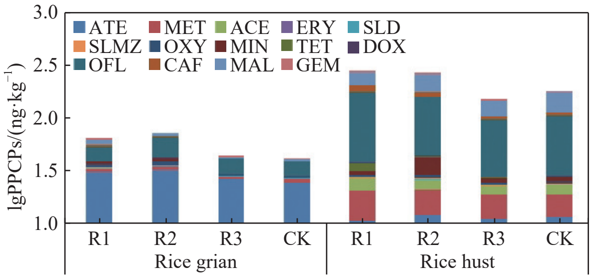


Figure 6 Changes of pharmaceutical and personal care products (PPCPs) content in rice grains and husks by different irrigation water sources and water level regulations.

oxytetracycline (OXY), metoprolol (MET) and minocycline (MIN), the PPCPs content was the largest of R2 (6.33 ng/kg), followed by R1 (6.82 ng/kg), R3 (5.00 ng/kg), and CK (4.79 ng/kg). Compared to CK, the PPCPs content of R1, R2 and R3 was increased by 32.3%, 42.4% and 4.4%, respectively. For rice husks, the high content of PPCPs mainly included ATE, OFL, acetaminophen (ACE) (not detected in grain), MAL, OXY, MET, MIN and tetracycline (TET) (not detected in grains), the change of PPCPs content was the largest of R1 (11.48 ng/kg), followed by R2 (11.30 ng/kg), CK (9.92 ng/kg) and R3 (9.31 ng/kg). Compared to R3, the PPCPs content of R1, R2 and CK was increased by 23.3%, 21.4% and 6.4%, respectively. The average of PPCPs content in rice grains was 5.73 *μ*g/kg, and that of rice husks was 10.50 μg/kg, namely the latter was 1.83 times of the former. In general, the PPCPs content in both rice grains and husks was at a very low level, but had a cumulative effect under irrigation of R1 and R2 water sources.

3.2 High efficiency of RDRW irrigation

3.2.1 Utilization of water and nitrogen

Utilization of water and nitrogen in paddy field under different irrigation water sources and water level regulations can be seen in Table 7. It showed that the yield can be effectively improved by RDRW irrigation. Compared to CK, the yield under R1, R2 and R3 was increased by 6.3%, 7.6% and 5.4% respectively. IWA under R3 and CK (R3≈CK) was relatively smaller than that of R1 and R2 (R1≈R2). The reason may be the one that the content of COD, nitrogen and organic matter in RDRW of R1 and R2 was relatively high, which affected the ventilation and leakage of paddy soil, thus resulting in large water consumption and slightly high IWA irrigated by R3 and CK. WCT, WUE_I and WUE_ET under R1 and R2 irrigation were slightly higher than those under R3 and CK irrigation. Meanwhile, RUE and NUE can be significantly improved by RDRW irrigation. Compared to CK, RUE and NUE of R1, R2 and R3 was increased by 6.7% and 24.2%, 9.3% and 22.6%, 9.4% and 21.7%, respectively. Under different water level regulations, yield, IWA, WCT, RUE and NUE were increased, but WUE_I and WUE_ET were decreased as the field water level increased. Compared to W1, yield, IWA, WCT, RUE and NUE were increased by –0.1%, 18.0%, 6.8% and 3.4% under W2, 2.1%, 30.6% 7.3% and 14.0% under W3, respectively, WUE_I and WUE_ET were decreased by 15.4% and 3.3% under W2, by 21.9% and 5.2% under W3, respectively. In addition, water level regulations had a significant impact on all indicators of utilization of water and nitrogen except WUE_ET, and irrigation water sources had a significant impact on yield, IRA, RUE and NUE, and interactions between water level regulations and irrigation water sources had a significant impact on yield, RUE and NUE.

Table 7 Utilization of water and nitrogen in paddy field under different irrigation water sources and water level regulations

Treatment	Yield/kg·hm^{-2}	IWA/mm	WCT/mm	WUE_I/kg·m^{-3}	WUE_ET/kg·m^{-3}	RUE/%	NUE/%
W1R1	10021bcd	367.2ef	804.8e	2.73a	1.25ab	72.9ab	83.2de
W1R2	9813cd	356.1f	815.5e	2.76a	1.20abc	72.6ab	85.4cd
W1R3	10174b	405.5cd	823.5de	2.51ab	1.24ab	59.1d	81.2ef
W1CK	9152f	387.1de	815.5e	2.36ab	1.12abc	53.8e	72.3g
W2R1	10054bc	425.0c	841.7bcde	2.37ab	1.19abc	58.6 d	91.2b
W2R2	9564e	429.9c	824.5de	2.22ab	1.16abc	71.2ab	86.3cd
W2R3	10224b	462.4b	860.5abcd	2.21ab	1.19abc	75.2a	88.2bc
W2CK	9271f	471.9b	836.3cde	1.96b	1.11bc	71.0b	67.3h
W3R1	9920cd	492.5ab	871.8abc	2.01b	1.14abc	72.1b	96.5a
W3R2	10968a	478.0b	868.2abc	2.29ab	1.26a	64.8c	95.8a
W3R3	9340f	488.9b	877.5ab	1.91b	1.06c	74.4ab	96.2a
W3CK	9790d	519.6a	886.5a	1.88b	1.10abc	66.0c	78.6f
W	**(*p*=0.00)	**(*p*=0.00)	**(*p*=0.00)	**(*p*=0.00)	NS(*p*=0.18)	**(*p*=0.00)	**(*p*=0.00)
R	**(*p*=0.00)	**(*p*=0.00)	NS(*p*=0.32)	NS(*p*=0.12)	NS(*p*=0.08)	**(*p*=0.00)	**(*p*=0.00)
W×R	**(*p*=0.00)	NS (*p*=0.15)	NS(*p*=0.77)	NS(*p*=0.94)	NS(*p*=0.20)	**(*p*=0.00)	**(*p*=0.00)

IWA, WCT, WUE_I, WUE_ET, RUE and NUE represented irrigation water amount, water consumption, water use efficiency of irrigation, water use efficiency of crop water consumption, rainwater use efficiency and nitrogen use efficiency, respectively. W and R represented water level control and water sources of irrigation, respectively. The values followed by the same letter in the column were not significantly different at a probability level of 0.05. NS meant not significant (p>0.05), * meant p<0.05, and ** meant p<0.01

3.2.2 Microbial diversity in root layer of paddy field

In this paper, the effective tags of all samples were clustered, and the sequence was clustered into operational taxonomic units (OTUs) with 97% identity. At the same time, the diversity of microbial community was reflected by alpha diversity (Table 8). OTUs reflects species diversity, Shannon index reflects community diversity, and Chao1 index reflects community richness. Under different irrigation water sources, it showed that the peak values of OTUs, Shannon and Chao1 were appeared under R3 irrigation.

Table 8 Operational taxonomic units (OTUs) number and alpha diversity indices

Sample	Total tags	OTUs	Shannon	Chao1
W1R1	42 774de	1775e	8.609de	2218.349gh
W2R1	50 321a	1784e	8.245e	2009.174h
W3R1	45 546b	2652b	9.569a	3249.588bc
W1R2	44 407bcd	3076a	9.611a	3707.506a
W2R2	40 950e	2302c	7.586f	2880.793de
W3R2	42 654de	2075d	9.065bc	2320.385fg
W1R3	45 324bc	3102a	9.632a	3125.846c
W2R3	44 528bcd	3215a	9.375ab	3372.359b
W3R3	45 680b	3084a	8.937cd	3098.724cd
W1CK	43 052cde	2429c	8.504e	2756.360e
W2CK	44 253bcd	2407c	8.464e	2883.915de
W3CK	43 885bcd	1978de	8.343e	2483.521f
W	NS(*p*=0.13)	**(*p*=0.00)	**(*p*=0.00)	**(*p*=0.00)
R	**(*p*=0.00)	**(*p*=0.00)	**(*p*=0.00)	**(*p*=0.00)
W×R	**(*p*=0.00)	**(*p*=0.00)	**(*p*=0.00)	**(*p*=0.00)

Except for R1 irrigation, the indexes of OTUs, Shannon and Chao1 of R2 and R3 were higher than those of CK, indicating that the species diversity, community diversity and richness of root layer soil under RDRW irrigation were higher than those of CK. Under different water level regulations, all indexes values were largest under W1, followed by W3 and W2, indicating that species diversity, community diversity and richness were decreased with the increase of field water level control. In addition, the effects of irrigation water source, water level regulation and their interactions on microbial diversity in paddy field were extremely significant.

Shannon and Chao1 represented the indicators of alpha diversity. W and R represented water level control and water sources of irrigation, respectively.

The values followed by the same letter in the column were not significantly different at a probability level of 0.05. NS meant not significant (p>0.05), * meant p<0.05, and ** meant p<0.01

3.3 Safe and efficient regulation mechanism of RDRW irrigation

Nine indexes were selected as the evaluation factors to establish the initial matrix, related to soil and crop safety (potential ecological risk index RI and PPCPs content in soil, content of heavy metals and PPCPs in grain), efficient utilization of water and nitrogen (WUE$_\mathrm{I}$, RUE, and NUE), soil microbial diversity (Shannon index), and economic benefit (yield). According to the principle that the larger the RUE, NUE, Shannon and yield are, the smaller the WUE$_\mathrm{I}$, RI and PPCPs content in paddy field and heavy metals and PPCPs content in rice grain are, the better the regulation schemes are, thereby the standardized decision matrix can be constructed. The entry weight W (w1, w2, w3, w4, w5, w6, w7, w8, w9) was calculated to be equal to 0.157, 0.099, 0.087, 0.175, 0.107, 0.086, 0.086, 0.084, and 0.119. The ideal solution x^+ and negative ideal solution x^- was determined, as x^+ = (0.0000, 0.0000, 0.0000, 0.0864, 0.0862, 0.0836, 0.1190), x^- = (0.1574, 0.0989, 0.0870, 0.1750, 0.1065, 0.0000, 0.0000, 0.0000, 0.0000, 0.0000). The Euclidean distance of ideal solution and negative ideal solution (di^+ and di^-), and the relative closeness of ideal solutions (S_i) under different regulation schemes were respectively calculated, and the results were ranked from large to small (Table 9). As it was shown, the order of comprehensive benefits from superior to inferior was R2, R1, R3 and CK under different water sources, and it was W3, W1 and W2 under different water level regulations, indicating that the high water level regulation with R2 water source irrigation was conducive to the exertion of the comprehensive benefits of RDRW irrigation regulation. Therefore, in the process of RDRW irrigation and reuse regulation, R2 was given as the priority water source, following R3 and CK as the supplementary water sources.

Table 9 Distance of ideal solution (d_i^+), negative ideal solution (d_i^-) and the relative closeness of ideal solutions (S_i) of different irrigation water sources and water level regulations

Scheme	d_i^+	d_i^-	S_i	Sequencing
W1R1	0.2504	0.1274	0.6628	3
W1R2	0.2955	0.1182	0.7143	2
W1R3	0.1447	0.2618	0.3560	9
W1CK	0.1129	0.2798	0.2876	12
W2R1	0.2235	0.1451	0.6064	5
W2R2	0.2596	0.1737	0.5992	6
W2R3	0.1540	0.2707	0.3627	7
W2CK	0.1265	0.2846	0.3077	9
W3R1	0.2540	0.1337	0.6551	4
W3R2	0.2953	0.1124	0.7244	1
W3R3	0.1335	0.3033	0.3056	11
W3CK	0.0961	0.2770	0.2575	13

3.4 Effectiveness evaluation in demonstration area

The rice planting area in the demonstration area is about 10 hm^2. The field irrigation was regulated by W3, and R2 was given as priority water source, while R3 and CK were used as supplementary water sources during the rice growth stages. Effectiveness indicators of crop and soil system in rice planting demonstration area are listed in Table 10. It showed that the irrigation water amount was 635 mm, including 215 mm of R2 water source, 300 mm of R3 water source, and 120 mm of CK. Compared to clear water irrigation, the consumption of fresh water can be reduced by 530 mm. For paddy field, the Cd content was remained unchanged, the content of Pb, Cr and PPCPs was increased slightly, with an increase of 0.3%, 0.9% and 2.5%, respectively. For rice grain, the Pb content was decreased, the Cd content was remained unchanged, and the content of Cr and PPCPs was increased slightly, with an increase of 1.3% and 1.5% respectively, and rice yield increased by 9.6%. It indicated that, on the basis of ensuring the safety of soil and crops, the consumption of fresh water can be significantly reduced, and the increase benefit of yield was significant.

Table 10 Effectiveness indicators of crop and soil system in rice planting demonstration area

Before and after irrigation	Irrigation water amount/mm			Heavy metals in soil/mg·kg^{-1}			PPCPsin soil/μg·kg^{-1}	Heavy metals in grain/mg·kg^{-1}			PPCPs in grain/ng·kg^{-1}	Yield/kg·hm^{-2}
	R2	R3	CK	Cd	Pb	Cr		Cd	Pb	Cr		
Clear water irrigation	/	/	650	0.05	33.23	24.36	0.922	0.017	0.115	0.715	4.78	9875
Reclaimed irrigation	215	300	120	0.05	33.31	24.78	0.945	0.017	0.114	0.724	4.85	10 824

4 Discussion

4.1 Mechanism on changes of heavy metals and PPCPs

The pollution degree of heavy metals is related to the source of reclaimed water, soil type, irrigation regulation mode and so on. The accumulation of heavy metals in soil will directly affect growth and quality of crop. Reasonable irrigation methods can reduce the accumulation of heavy metals in soil. Li et al.[19] proposed that drip irrigation or alternate root irrigation technology can significantly reduce the content of heavy metals in soil compared with conventional irrigation. In this paper, field water level regulations were carried out for rice, and it was found that water level regulation had a significant impact on Cd content in both 0-20 cm and 20-40 cm soil layers in paddy field, which further confirmed the effective effect of irrigation regulation on the accumulation and distribution of heavy metals. Li et al.[20] found that there were great differences in the accumulation of heavy metals in various parts of crops, and the lowest one in grains, whereas the rhizome leaf system played a barrier role in the migration of heavy metals to fruits, which was basically consistent with the research conclusions of this paper. Xiao et al.[21] analyzed the correlation between soil heavy metal content and soil chemical properties under reclaimed water irrigation, with the result showing that Cd was significantly correlated with soil EC, Pb was significantly correlated with OM (organic matter) and TN (total nitrogen), and Cu was significantly correlated with NH_4^+-N. Combined with the results in this paper, RDRW irrigation, on the one hand, affect the physical and chemical properties of the soil, on the other hand, it brought in nitrogen and other nutrients to affect the migration and distribution of soil nitrogen, and at the same time, it affected the soil biodiversity, that further affected the migration and accumulation of heavy metals. Although PPCPs content is at a continuously low level in the reclaimed water system, it will still lead to significant ecotoxicity. Gottschall et al.[22] monitored more than 80 kinds of PPCPs in groundwater and soil after one year of sludge application in the drainage network and activated sludge in Ottawa, Canada. Seven kinds of PPCPs in the drainage network, four kinds of PPCPs in groundwater and five kinds of PPCPs in soil were detected, respectively. Wang et al.[23] studied the impact of long-term reclaimed water irrigation on antibiotics in green land, and five tetracycline antibiotics and nine metabolites were detected from soil, and the total detected concentration of antibiotics was 12.7-145.2 μg/kg. The irrigation water source of these studies is basically urban sewage, while the irrigation water source of this study is rural domestic sewage, and the results showed that the PPCPs content in paddy field and rice plant irrigated by R1 water source was enriched, followed by R2, and the irrigation effect of R3 water source was basically consistent with CK. Therefore, in order to reduce the pollution risk of PPCPs, irrigation directly by R1 water source should be avoided, and R3 water source was relatively safe as a supplementary water source.

4.2 Effects mechanism on water-nitrogen use and soil microbial diversity

Reclaimed water irrigation changes the process of water and fertilizer absorption and utilization of water-nitrogen for crops. On the one hand, it changes the water-soluble salts in soil, and physically hinders water and fertilizer absorption. On the other hand, it affects soil environmental quality, causes change on soil permeability[24], affects indirectly the process of crop metabolism and water and fertilizer absorption[25], moreover changes the activity and biodiversity of soil microorganisms, and affects the nutrient transformation process[26]. Cakmakci et al.[27] found that the water use efficiency under subsurface drip irrigation with reclaimed water was 28.2% and 99.4% higher than that under surface drip irrigation and furrow irrigation. In this study, WUE_I and WUE_{ET} showed a downward trend under RDRW irrigation, and the main reason for this difference from Cakmakci's research is that the core of irrigation and drainage regulation adopted is to maximize the consumption of reclaimed water, rather than achieving the purpose of water-saving irrigation. Reclaimed water is rich in nitrogen, phosphorus and organic matter, with proper regulation of water and nitrogen it can improve soil buffering performance and soil fertility and promote crop growth[28,29]. The research in this paper showed that the rural domestic sewage regenerative irrigation can increase production by 6.7%-9.4%, and improve biodiversity, which effectively confirmed the above conclusions. The NH_4^+-N concentration of RDRW was from 3.52 to 11.9 mg/L, the nitrogen accumulation brought in by reclaimed water can reach 10.43-27.90 kg/hm^2, and NUE can be increased by 21.7%-24.2%, which provided a basis for fertilizer nitrogen application strategy of reclaimed water irrigation. Han et al.[30] found that it can save 75% of clean water amount with reclaimed water irrigation, and substitution efficiency of nitrogen fertilizer was about 35.8%. These research conclusions fully showed that reasonable irrigation regulation of reclaimed water can improve nitrogen availability and utilization, and promote crop yield[31]. In this paper, three kinds of field water level regulations were set up, in addition to the drainage affected by typhoon, there was basically zero direct drainage, which effectively improved the utilization efficiency of reclaimed water, and reduced the amount of fresh water up to 2000-6000 m^3/hm^2.

5 Conclusions

This study considered the interaction of soil and crop safety, utilization efficiency of water and nitrogen, soil microbial diversity, and the innovation lied in the establishment of an efficient and safe regulation mechanism of reclaimed water irrigation in paddy field was established.

For field experiments, under RDRW irrigation, the content of Cd and Pb was increased slightly, while the content of Cr, Cu and Zn was decreased, the ecological risk was mild, RI under R3 water source irrigation was lowest in paddy field, the heavy metals content in rice grains did not increase significantly, which met the requirements of pollutant limit in the rice. The PPCPs contents in soil were higher than that of CK, accumulated in 60-80 cm soil layer, but PPCPs contents in rice husks and grains were at a very low level. Compared to CK, the yield, RUE and NUE was increased by 5.4%-7.6%, 6.7%-9.4%, and 21.7%-24.2%, respectively, meanwhile, with the increase of field water level control, yield, IWA, WCT, RUE and NUE showed a significant upward trend. The soil species diversity, community diversity and richness was improved, but showed a downward trend with the increase of field water level control. The regulation of high water level control (W3) with R2 water source irrigation was conducive to the exertion of comprehensive benefits, R3 and CK can serve as the supplementary water sources.

For monitoring of the demonstration area, the consumption of fresh water was reduced by 530 mm, yield was increased by 9.6%, Pb, Cr and PPCPs content in paddy field was slightly increased by 0.3%, 0.9% and 2.5%, respectively, and Cr and PPCPs content in rice grains was slightly increased by 1.3% and 1.5%, respectively. Short-term irrigation of RDRW did not cause heavy metals and PPCPs pollution on soil and crops, not affecting crop yield, and it can meet the requirements of efficient and safe reuse of reclaimed water irrigation. However, the impact of long-term reclaimed water irrigation is not clear, so it is still necessary to track and monitor the effect on the system of soil, crop, and underground water.

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