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The Effect of Multi-Years Reclaimed Water Irrigation on Dryland Carbon Sequestration in the North China Plain

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Abstract: Reclaimed water is an alternative water source which could alleviate the shortage of water resources in agricultural systems. Many researchers have studied the effect of reclaimed water on soil environment, crop yield, etc. However, carbon sequestration in reclaimed water irrigated agricultural systems is less studied. This study investigates methane uptake and photosynthesis in reclaimed water irrigation systems contributing to carbon sequestration estimation and analyzes the important factors impacting them. The results show that CH_4 uptake is related to soil water-filled pore space (WFPS) with a quadratic and it has the highest uptake when WFPS is between 40 and 50%. Long-term reclaimed water irrigation could significantly decrease ($p < 0.05$) CH_4 uptake and macroaggregate stability in the topsoil. However, reclaimed water had no significant impact on photosynthesis in comparison. The type of fertilizer is an important factor which impacts CH_4 emission from soil; urea had a lower CH_4 uptake and a higher CO_2 emission than slow-released fertilizer. Overall, reclaimed water irrigation could effectively decrease soil carbon sequestration. A soil wetted proportion level of 40–50% was recommended in this study for favorable methane oxidation. Slow-released fertilizer in reclaimed water irrigated agriculture could better control soil carbon emission and soil carbon absorption.

Keywords: CH_4 uptake; photosynthesis; carbon exchange; soil environment; water-filled pore space; types of fertilizer

1. Introduction

Agricultural CH_4 production from soil plays an important role in global radiation and the greenhouse effect. Some studies showed that CH_4 contributes 18% of global radiation [1,2], and its greenhouse effect capacity is 26 times that of CO_2 [2]. Lashof et al. [3] and Jain et al. [4] considered that global CH_4 emissions were rising. Dryland carbon sequestration is necessary for global carbon balance, which includes CH_4 uptake and photosynthesis. Most studies showed that dryland CH_4 uptake accounts for 6% of the total global methane consumption, and it is a very important part of CH_4 sink [5,6]. The soil carbon sequestration could be impacted by many factors, such as temperature, soil water content, fertilizer, soil environment, etc. Temperature can significantly promote the activity of methane oxidizing bacteria and increase methane production [7]. Soil moisture can control bacteria activity and affect the soil oxygen content. Previous literature also studied the relationship between CH_4 oxidation capacity and soil moisture, showing that it increases with the increase in soil moisture [8]. Previous studies also demonstrated that there is a strong relationship between methane production and soil oxygen concentration [9]. Methane oxidizing bacteria could have a strong relationship with soil EC, nitrogen [10]. These driving factors can be divided into two types: (1) Dynamic type, water change and temperature could drive CH_4 emission and significantly impact crop growth. (2) Biochemistry type, that attributed soil properties could participate in CH_4 production processes and crop growth by impacting the soil microorganism and the soil electronic change.

In recent years, with the development of intensive agricultural systems, the water resources in some regions are vulnerable [11]. Reclaimed water as an alternative water source has gradually been used in agricultural irrigation. Most studiesshowed that long-term reclaimed water irrigation could effectively impact the soil pH, EC, and soil organic carbon [12,13] and could effectively impact crop growth and photosynthesis [14,15]. Moreover, some researchers showed that sewage water could increase soil CH_4 emission in the paddy field agricultural system [16,17]. These studies suggest that the increasing gas emissions from soil could contribute to the microorganism activity [18] or the changing soil environment [19] impacted by reclaimed water. However, few studies have explored the reason for this. Therefore, it is necessary to determine the effect of reclaimed water on soil CH_4 emission and observe the patterns of soil carbon sinks under reclaimed water irrigated agricultural systems. This effectively guided the reclaimed water irrigation and balanced carbon emission.

The present study focused on the effect of reclaimed water on the patterns of main carbon sequestration (dryland CH_4 uptake, crop photosynthesis) under two types of fertilizer (urea and slow-released fertilizer) and illustrated the factors affecting these changes under reclaimed water irrigation. Our findings could effectively guide the reclaimed water irrigation technology contributing to carbon balance in the soil–atmosphere system, and are beneficial to fertilizer in reclaimed water irrigated agriculture.

2. Materials and Methods

2.1. Experiment Design

The experiment site of Tongzhou, Beijing, China, is shown in Figure 1. The details of trail conditions and agricultural management are described in Chi et al. [20]. The experiment set two types of water quality (reclaimed water and underground water) and two types of fertilizer (urea and slow released fertilizer), as shown in Table 1. The properties of the soil in all treatments were measured at the beginning of the experiment. The sampling took place in October 2013 and the details are shown in Table 2.

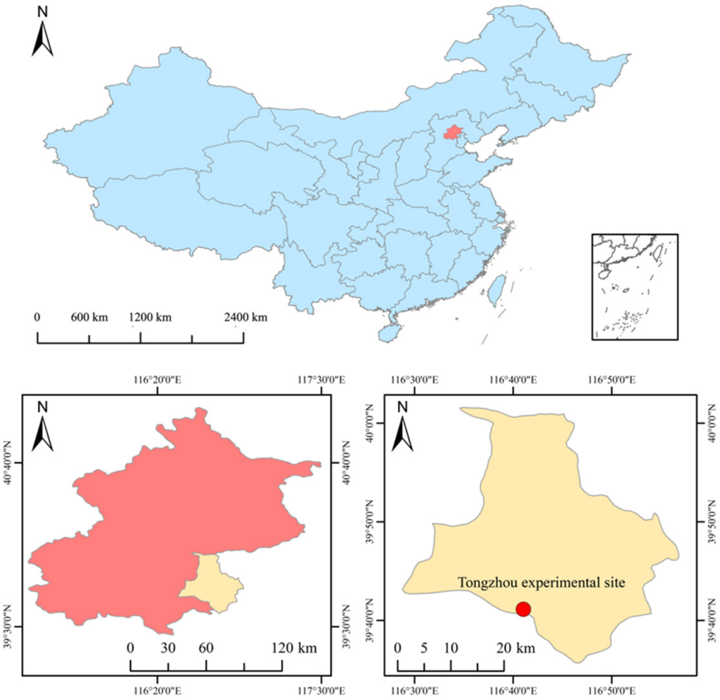

Figure 1. Location of experimental site.

Table 1. The number of experiment.

Item	Water Quality	Fertilizer	Number	Remarks
1	underground water/CW	urea/UF	CUF	control group
2	underground water/CW	slow released fertilizer/AF	CAF	
3	reclaimed water/RW	urea/UF	RUF	experimental group
4	reclaimed water/RW	slow released fertilizer/AF	RAF	

Table 2. The soil background value at beginning of planting.

Item		NO_3 (mg/k)	$NH_4{}^+$ (mg/kg)	pH	EC (us/cm)	TN (mg/kg)	SOC (mg/kg)
RUF	2013	8.21 ± 2.12	8.94 ± 2.12	7.81 ± 0.32	531.23 ± 12	3.12 ± 0.22	11.06 ± 0.13
CUF	2013	8.10 ± 1.94	8.73 ± 2.34	7.93 ± 0.11	654.51 ± 34	3.01 ± 0.22	8.01 ± 0.77
RAF	2013	8.11 ± 0.14	8.19 ± 3.42	8.01 ± 0.15	431.43 ± 24	2.94 ± 0.13	10.34 ± 0.54
CAF	2013	8.13 ± 2.34	8.41 ± 2.32	8.04 ± 0.24	351.71 ± 15	2.01 ± 0.14	11.23 ± 0.54

2.2. The Sampling Method and Collection

Soil CH_4 emission: The CH_4 emission from the soil was measured by chamber-gas chromatography, using Agilent GC-6820 (Agilent Technologies Inc., Santa Clara, CA, USA), 20 mL of gas was collected in each sample. The square chamber was 50 cm×50 cm×50 cm, and it was inserted to a depth of 5 cm in the soil. One chamber and stopwatch was inserted into each plot, 2–3 people sampled at the same time, and a gas collection was fulfilled in 10 s. Samples were taken between 14:00 p.m. and 16:00 p.m. in wheat growth season and between 9:00 a.m. and 11:00 a.m. in maize growth season [21]. Every treatment had three replicates. The calculation of CH_4 emission was described by Hashimoto et al. [22] and Konda et al. [23]. The soil average temperature of 0–20 cm was recorded when the gas was collected.

Crop photosynthesis: The rate of crop photosynthesis was measured by a portable photosynthetic gas analysis system (CI-340, CID, America), every treatment had 5 replicates. During the winter wheat period, the photosynthesis of flag leaf was measured. The sample was collected on early jointing, mid jointing, botting, heading, and filling stage. During the summer maize period, the collecting time was V3 (third leaf), V6 (sixth leaf), V12 (twelfth leaf), VT (tasseling), R2 (blister stage) and R3 (milk stage).

Carbon balance means to the carbon gas exchange. The calculation is as follows:

$$\text{Carbon balance} = C_4 + C_p - C_2$$

C_2 was cumulative CO_2 emission ($t.hm^{-2}$); C_p was et photosynthesis in wheat-summer rotation system ($t.hm^{-2}$); C_4 was cumulative CH_4 emission ($t.hm^{-2}$) ($hm^2 = 1000 \times m^2$), the value converted to CO_2-eq from soils during one crop production in wheat-summer rotation system.

WFPS and temperature: soil water content and temperature were measured by soil moisture and temperature sensors (ET-100, Insentek, China), the buried depth of sensor was 1 m. The WFPS in 20 cm depth soil was as follows:

$$\text{WFPS} = \frac{\theta_v}{1 - r/\rho} \quad (1)$$

θ_v was the soil water content; r was the soil bulk density; ρ was the soil density 2.65 g/cm^2.

pH, EC and soil nitrogen: Soil samples were collected when the photosynthesis was measured, every treatment had five replicates and the depth was 0–30 cm. The pH and EC was measured by a multi-parameter tester (SG23, Mettler Toledo, Shanghai, China) and the soil nitrogen ($NH_4{}^+$, $NO_3{}^-$) was measured by a continuous flowing analyzer (Alliance FUTURA, AMS, Frépillon, France). The measurement method of soil properties referred to can be found in [24]. Soil particle size was measured by a laser particle sizer (0.01–3500 um, Mastersizer 3000, Malvern, England).

2.3. Data Analysis

All statistical analyses were carried out using SPSS V26 for MacOS (SPSS Inc., Chicago, IL, USA). Analysis of variance (ANOVA) was used to determine treatment effects. The least significant difference procedure (LSD) and contrasts with a probability level of 0.05 were used to determine significant differences between treatment means by using a t-test. In this study, average refers to the arithmetic mean. Correlations were assessed by Pearson's correlation coefficient (α). The soil carbon exchange in this paper refers to the carbon content exchanged between soil and atmosphere during the main planting period in winter wheat and summer maize rotation.

3. Results

3.1. Principal Component Analysis

The PC1 was 55.1% and the PC2 was 24.9%. As shown in Figure 2, UF treatments in PC2 were higher than AF treatments, and RW (reclaimed water) treatments in PC1 were higher than CW (clean/underground water) treatments. Type of fertilizer respect PC2, type of water respect PC1. EC, NO_3^-, N_2O and NH_4^+ occurred on the first quadrant. pH, SOC and CO_2 occurred on the fourth quadrant. CH_4 was at the second quadrant. The N_2O emission flux was related to EC, NO_3^-, NH_4^+ and TN, the CO_2 emission flux was related to SOC, pH and TN, but these soil properties had no relationship with soil CH4 emission. Moreover, all treatments were divided into two parts according to fertilizer type (Figure 2). During the four-year experiment, reclaimed water could effectively soil pH, TN, NO_3^-, and the greenhouse gas (CO_2, N_2O and CH_4) was related to the type quality of irrigated water. The absolute value of NH_4^+, EC and SOC in PC2 was higher than PC1.

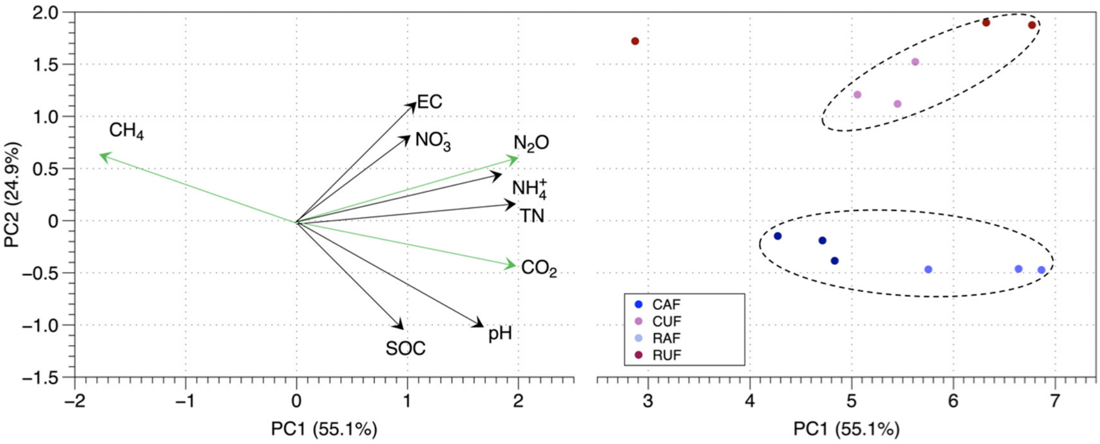

Figure 2. The patterns of soil temperature and WFPS from 2014 to 2015.

3.2. Soil CH_4 Emission

As shown in Figure 3, soil could absorb methane effectively from 2013 to 2014, and the patterns of methane emission flux were similar in all treatments. As shown in Table 3, reclaimed water could significantly decrease methane uptake between 11.76 and 27.27%, AF could increase the methane uptake in comparison with UF. The order of methane cumulative uptake was RUF < RAF < CUF < CAF.

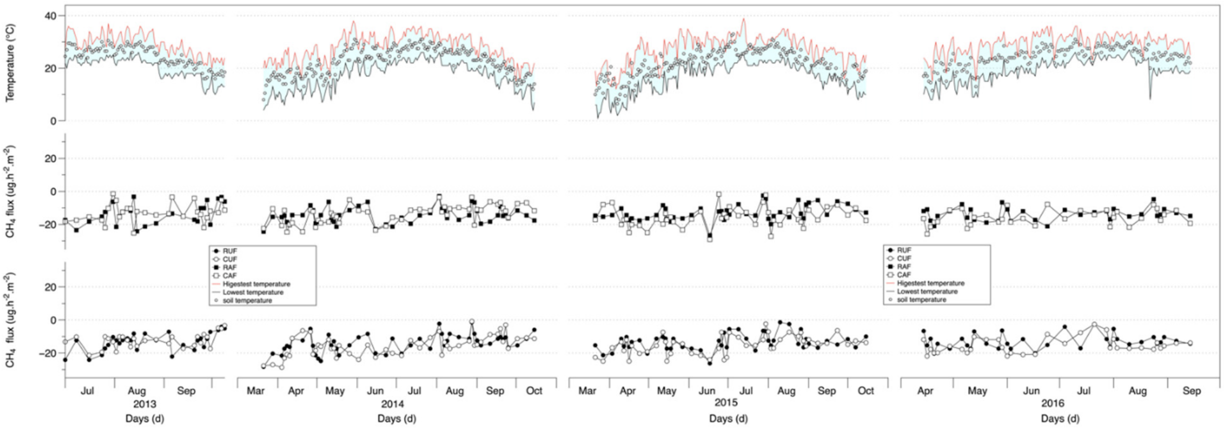

Figure 3. The patterns of temperature and soil CH_4 emission flux from 2013 to 2016.

Table 3. The average and cumulative CH_4 emission from 2013 to 2016.

Treatment	2013		2014		2015		2016	
	Aver	Cum	Aver	Cum	Aver	Cum	Aver	Cum
RUF	−13.21 a	−0.079 a	−13.79 a	−0.17 a	−13.25 a	−0.16 a	−12.87 a	−0.11 a
CUF	−12.47 a	−0.074 a	−15.26 b	−0.19 b	−15.04 b	−0.18 b	−14.86 b	−0.14 b
RAF	−13.53 a	−0.081 a	−13.08 a	−0.16 a	−13.63 a	−0.17 a	−13.45 a	−0.12 a
CAF	−12.93 a	−0.077 a	−13.14 a	−0.17 a	−15.58 b	−0.19 b	−15.12 b	−0.14 b

Aver means the average CH_4 emission flux during the growth period, unit is ug.h^{-1}.m^{-2}.; Cum means the cumulative CH_4 emission flux during the growth period, unit is t.hm^{-2}. a.b indicate that the significant analysis between different water quality types within the same fertilizer. Means followed by the same letter are not significantly different according to t-test ($p < 0.05$).

In order to analyze the factors that could impact methane emission, the paper shows the relationship between methane and WFPS, soil nitrogen. As shown in Figure 4, the relationship between WFPS and methane was quadratic, and the R^2 in AF treatment was higher UF. Meanwhile, the RAF (0.15) and RUF (0.34) was higher CUF (0.13) and CAF (0.26). As shown in Figure 4, there was a strong linear relationship between methane and soil NH_4^+, the R^2 in RUF and CUF was 0.65 and 0.32, respectively (Table 4). However, there is no relationship between methane and soil NO_3^-.

Table 4. The regression between CH_4 emission and soil WFPS, NH_4^+ content.

The Regression		Equation	R^2
Y-CH_4 Emission	x		
RUF	WFPS	$Y= -30.82 + 53.5x - 36.97x^2$	0.15
RAF	WFPS	$Y = 51.00 - 267.03x + 271.82x^2$	0.34 **
CUF	WFPS	$Y= 31.84 - 228.92x + 266.67x^2$	0.13
CAF	WFPS	$Y= 26.55 - 218.92x + 267.83x^2$	0.26 **
RUF	NH_4^+	$Y= -21.89 + 0.96x$	0.65 *
CUF	NH_4^+	$Y= -19.45 + 0.68x$	0.32

* indicates that $p < 0.05$ according to T-TEST test; ** indicates that $p < 0.01$ according to T-TEST test.

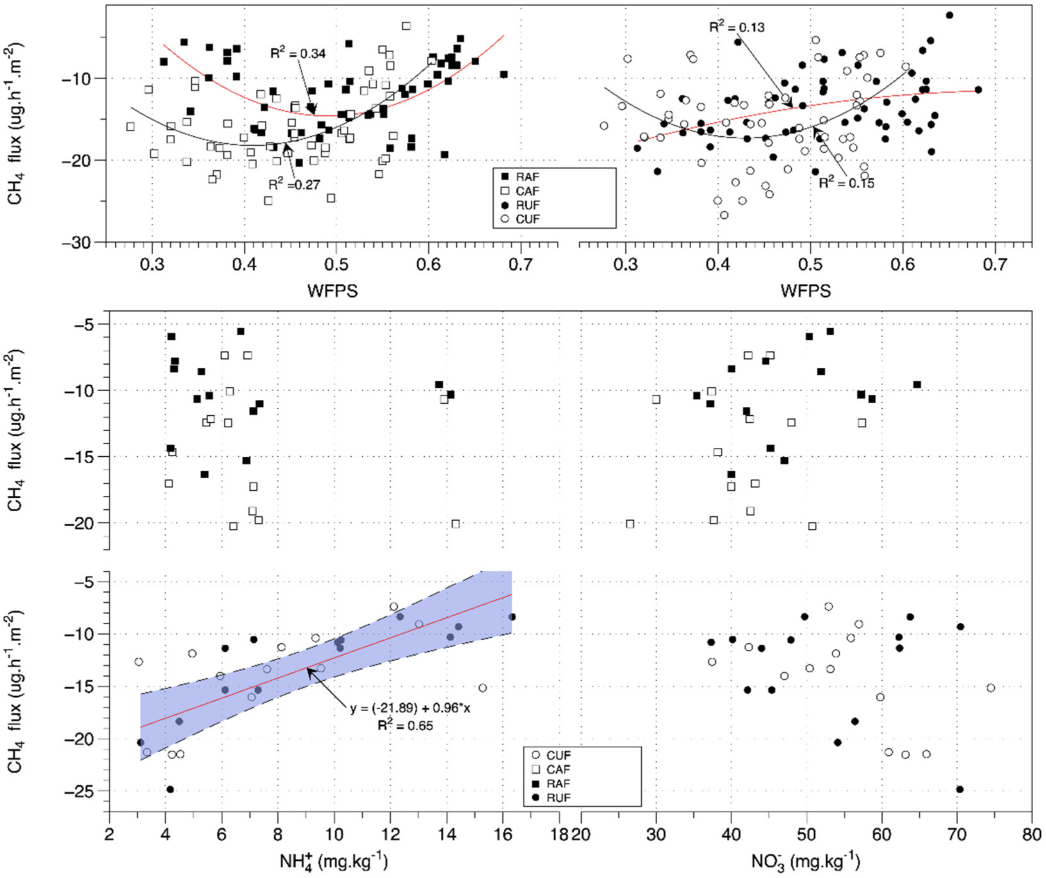

Figure 4. The regression between soil CH_4 emission and WFPS, NH_4^+ and NO_3^-. The shaded area is 95% confidence interval.

3.3. Photosynthesis

As shown in Figure 5, photosynthesis from 2014 to 2015 had similar varieties among all treatments; photosynthesis first increased and then decreased. There is a strong correlation between photosynthesis and temperature, the highest value appeared between 12:00 and 15:00, and the average value in the summer-maize period was higher than in the winter-wheat period. The highest value appeared in August during the summer-maize period, white-wheat kept it steady among all treatments. As shown in Table 5, there was no significant difference ($p > 0.05$) between RW and CW, and photosynthesis in UF was higher than AF during the whole period.

Table 5. Cumulative carbon balance calculation from 2014 to 2015.

Treatment	CO_2 Emission t.hm^{-2}	CH_4 Emission t.hm^{-2}	Photosynthesis t.hm^{-2}	Carbon Balance t.hm^{-2}
RAF	64.41 a	−0.31 a	119.00 a	−54.90 a
RUF	66.61 a	−0.33 a	108.39 a	−42.11 a
CAF	61.64 a	−0.37 b	115.80 a	−54.53 a
CUF	62.51 a	−0.36 b	112.11 a	−49.96 a

CO_2 emission from 2014 to 2015 was listed on Table 4 refer to [22]; CO_2 emission and CH_4 emission is accumulative gas emission from 2014 to 2015. photosynthesis is average photosynthesis multiply time. Means followed by the same letter within a column are not significantly different according to t-test ($p < 0.05$).

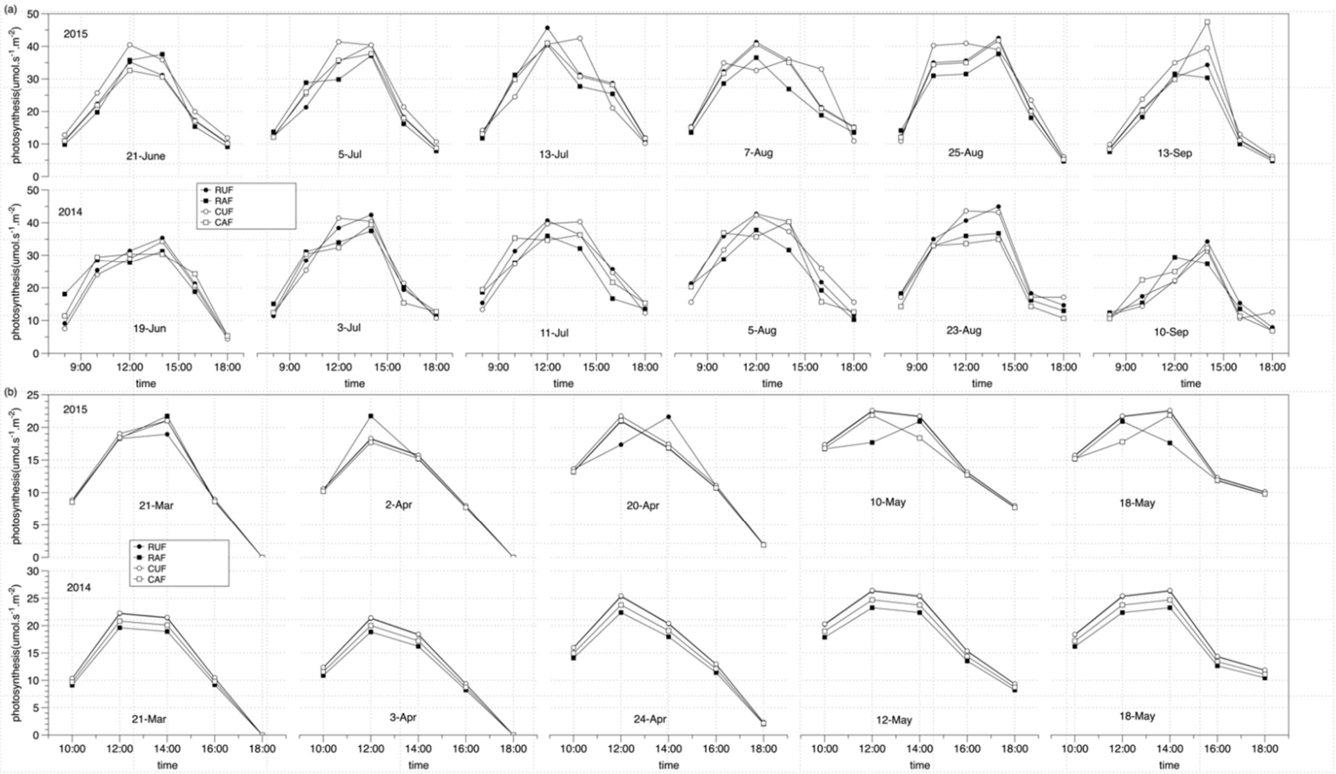

Figure 5. (**a**) The varieties of photosynthesis during the summer-maize period from 2014 to 2015; (**b**) The varieties of photosynthesis during the winter-wheat period from 2014 to 2015.

3.4. The Effect of RW on Soil Carbon Balance

CH_4 uptake in dryland and plant photosynthesis were necessary parts of carbon sink. As shown in Table 5, the paper has demonstrated that RW can significantly decrease soil CH_4 emission and not affect plant photosynthesis. Although RW could increase the CO_2 emission in comparison with CW, RW could not impact carbon balance during the whole growth period in 2014 and 2015. Facing the field carbon balance, fertilization and field management were the key factors to determine the difference. By contrast, reclaimed water could effectively increase CO_2 emission and decrease soil CH_4 uptake, but it had no effect on carbon exchange between farmland and atmosphere.

3.5. The Effect of RW on Soil Particle Size

As shown in Figure 6, the distribution of soil particles was mainly attributed to the quality of irrigation water. Urea and slow-released fertilizer had no significant ($p > 0.05$) effect on the distribution. The soil particle size of RW was significantly ($p < 0.05$) larger than that of CW between 10–100 um. When soil particle size is greater than 10 um, the distribution in CW was significantly ($p < 0.05$) higher than RW. When soil particle size was less than 1 um, there was no significant difference between RW and CW from 2014 to 2015.

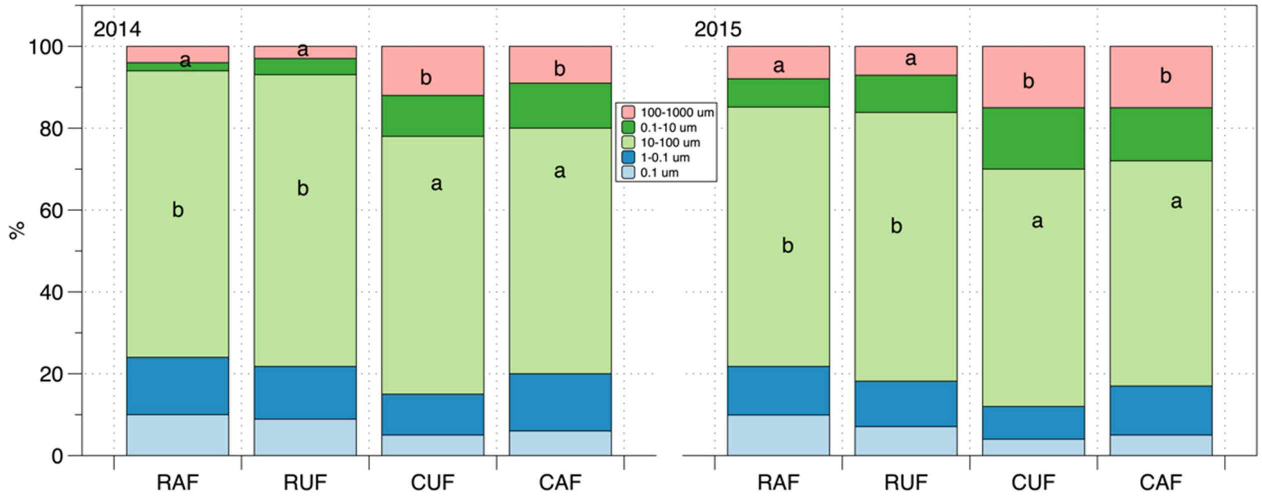

Figure 6. The distribution of particle size among all treatment in 2014 and 2015. Means followed by the same letter within a particle size range are not significantly different according to t-test ($p < 0.05$).

4. Discussion

In this paper it is demonstrated that methane is absorbed and its uptake has a strong relationship with WFPS. Other studies also considered that WFPS is an important factor which impacts CH_4 uptake [25]. This could largely be attributed to the oxygen content in soil; the production of soil CH_4 is driven by soil microorganism, methane oxidizing bacteria [26]. Some researchers showed that methane oxidizing bacterial reproduction could be provided by soil nutrients [27] or a good oxygen environment [28,29]. Although, in this study, reclaimed water could significantly decrease methane uptake from soil (Table 3), there was no difference between reclaimed water and clean water in soil nitrogen, pH, and other factors (Figure 1). Therefore, RW might affect the distribution of soil particle size to impact soil aeration as that is the main factor affecting soil methane emission. Macroaggregate stability decreased for reclaimed water in our study (Figure 5) and soil organic carbon and soil nitrogen (Table 2). Some studies showed that a lower soil gas diffusivity and a higher mineral N content in soil could significantly decrease CH_4 uptake [30]. Some researchers considered that reclaimed water irrigation could cause soil hardening and the soil structure quality could decrease with increasing sewage irrigation years [31,32]. Our results illustrated that RW had a better soil aeration in comparison with CW, and soil aeration in RW could be easily impacted by soil WFPS. As shown in Table 3, the correlation R^2 in RW was higher than CW. DZ et al. [33] also consider that reclaimed water irrigation could decrease soil water holding capacity and increase soil penetration resistance.

However, soil respiration is also necessary to impact CH_4 uptake. The increasing soil respiration could effectively decrease CH_4 emission from soil [34]. Liang et al. [35] showed that reclaimed water could improve soil respiration. Therefore, reclaimed water decreases CH_4 uptake. It also explains the difference between UF and AF in CH_4 uptake. Soil microbial activity could be affected by fertilizer type [36] and it could impact the activity of aerobic methanotrophs. As shown in Table 3, the R^2 in AF was higher UF in addition to the effect of fertilization. The difference between them is attributed to the fertilizer method, urea could raise significantly soil respiration during the 3–5 days after fertilization [37,38], it could accelerate the formation of tiny anaerobic areas in soil porosity in comparison with AF treatment.

RW as an alternative water source could effectively impact soil properties, increase soil NO_3^- content [39], reduce soil pH, increase SOC [40] and EC. In this study, the carbon balance was calculated, and it was demonstrated that there is no significant ($p > 0.05$) difference between RW and CW. The variety of SOC increased only with the increase in irrigation and fertilization years (Table 1). However, the effect of reclaimed water irrigation on wheat and maize photosynthesis had no significant difference with clean water irrigation, as shown in Figure 4. Photosynthesis could be influenced by yield, illumination, soil microelement, etc. Most studies showed that reclaimed water could not significantly impact the yield [41]. Soil nitrogen, potassium, and magnesium play an important role in plant photosynthesis [42]. In our experiment, it is difficult to reflect on the effect of reclaimed water on crop photosynthesis because the amount of nitrogen applied is much greater than that in reclaimed water. In terms of carbon emission, RW could significantly improve soil respiration by increasing the abundance of microorganisms [43,44]. However, CO_2 emission not only includes microorganism activity, but also crop root respiration. Irrigation, fertilizer, and air environment play an important role in crop root respiration [45] and these are similar among all treatments.

5. Conclusions

Quadratic correlation ($p < 0.05$) between methane flux and WFPS was found in this study, and the level of 40%~50% WFPS had an advantage on methane oxidation. Irrigation, water quantity, and water quality are important factors in CH_4 uptake on dryland, although they did not significantly impact crop photosynthesis. In addition, long term irrigation with reclaimed water can significantly decrease macroaggregate stability to decrease CH_4 uptake 11.76%~27.27%. The application of urea enhanced CO_2 emission and decreased CH_4 uptake in comparison with the application of slow-release fertilizer, this is likely due to high levels of ammonium. Overall, slow-release fertilizer is recommended in this study in order to favorably control carbon gas emissions and to effectively increase carbon sequestration under reclaimed water irrigated agriculture.

Author Contributions: Y.C. and P.Y. designed the experiments; Y.C. and N.M. conducted the experiments.; N.M and Q.Z. contributed materials.; Y.C. and Q.Z. analyzed the data and wrote the paper.; S.R. revised the paper. All authors have read and agreed to the published version of the manuscript.

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Conflicts of Interest: The authors declare no conflict of interest.

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