Evaluation of Chemical Impacts of Rice Irrigated with Urban Treated Wastewater

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Abstract. Improving practices of water saving in irrigation is a priority facing global climate changes. To cope with water scarcity, the irrigation with treated wastewater (TWW) is becoming a solution, posing however several health and environmental risks on agricultural fields. This preliminary study aimed to assess the physicochemical impacts of rice irrigated with TWW applied with a subsurface drip irrigation (SDI) system, focusing the rice food, soil and environmental safety. Normal water and flooding was used as a reference for comparison. The experimental scheme considered three treatments with five repetitions, namely the TWW irrigation with SDI, the normal water with SDI, and the flooded irrigation with normal water. The pots filled with 15 L of soil, sowed with a local traditional rice variety, were kept outdoors. The fertilization scheme followed the usual one under field conditions. The irrigation frequency varied from three to five times a week. Measurements of physicochemical properties of irrigation water, drainage and rice grain were carried out, according to the analytical reference methods. Results showed that the irrigation with TWW rises the electric conductivity of drainage water, but that the rice grain does not present increased risks to public health due to the low content of arsenic, cadmium, lead and mercury. However, the irrigation method must be adapted to a SDI to avoid human and animal contact with putative contaminants present in this water and thus safeguarding Environmental and Food Safety. Long term soil effects of TWW, including winter percolation, will be assessed in the future.

Keywords: Rice irrigation · Chemical impacts · Subsurface drip irrigation · Treated wastewater

1 Introduction

Rice is the world most important food crop, and the staple food for more than half the human population. Its demand in the world market has increased, and it is foreseeable a

J. R. da Costa Sanches Galvão et al. (Eds.): *Proceedings of the 1st International Conference on Water Energy Food and Sustainability (ICoWEFS 2021)*, pp. 796–805, 2021.
https://doi.org/10.1007/978-3-030-75315-3_84

continuos rise around 24% over the next 20 years [1]. The growth of African and Asian populations, as well as the changes in diet, explain the increase of human rice consumption. In the Mediterranean countries it is cultivated on about 1.30 million hectares [2], mainly in flat alluvial regions, with a relative abundance of water. Traditionally, it is grown under continuous flooding.

To respond to the growing market demand, rice production can be increased enlarging its cultivation area; however, there are limitations hard to overcome. The main constrainment is the water supply, due to its unavailability, high costs in many regions, and for countering a demand from society to restrict its use in irrigation. Moreover, flooding is effective on flat, deep and poorly permeable soils, which are preciselly the soils where rice is traditionally cultivated; conditions that are scarce in other areas to allow the expansion of rice paddies. Many studies had explored the sustainability of innovative irrigation options, in order to reduce water consumption for rice production and its negative environmental impacts, and to extend rice cultivation outside traditional rice areas [3].

Nowadays, improving irrigation practices to save water is a priority. Therefore, the use of drip irrigation for rice cultivation has been one of the recommended solutions [3, 4], since it solves some of the most critical issues of rice continuous flooding, namely, it allows: i) reducing water consumption, especially the fraction of deep percolation; ii) the use of lower quality water, in terms of salinity and microbiology (high microbiological load in subsurfce to do pose direct risk to human and animal healty); iii) facilitating the automation of irrigation, reducing labor; and iv) to reduce the residual cadmium and arsenic contents in the seeds.

In this context, the use of treated wastewater (TWW) for irrigation is a solution to consider to deal with water scarcity. In certain places, TWW it is a significant source of water for irrigation with guaranteed supply during the summer, despite posing several health and the environmental risks [5].

The aim of this study was to evaluate the physicochemical impact of the rice irrigation with urban TWW on drainage water from soil percolation (environmental safety) and on rice grain quality (food safety). For this purpose some physicochemical quality parameters were monitored in the irrigation and drainage water, as well as the effect of the irrigation method on the rice grain quality.

2 Material and Methods

2.1 Experimental Layout

To assess the impacts of reusing municipal TWW for rice irrigation in Lis valley, the environmental exposition of drainage water to chemical coumpounds of TWW was determined through the physicochemical analysis of soil water samples; wheareas the rice consumer exposition to heavy metals was assessed through rice grain analysis.

Two sources of irrigation water (Treated WasteWater, TWW and Normal Water, NW, used for comparison) and two irrigation methods (Subsurface Drip Irrigation, SDI and Continuous Flooding, F) were used for this evaluation in a pot tests.

The experimental layout considered three treatments with five repetitions, namely: i) irrigation by SDI with TWW, ii) irrigation by SDI with NW, and iii) irrigation by continuous flooding with NW. The NW was collected from a well, whereas the Urban TWW was supplied by the Coimbrões WWTP, AdCL SA, Leiria. The pots with 15L of soil, seeded with a traditional variety of local rice (*Oryza sativa* L. cv Ariete), were kept outdoors. The fertilization scheme followed the usual field conditions. The irrigation frequency varied between three to five times a week for pots with SDI and flooding, respectively. The SDI with dripper at a depth of 15 cm, supplied through a small reservoir. A rice field owned and managed by a farmer, irrigated with NW, was used for comparison. Measurements of soil texture, irrigation water volume were recorded. Drainage water samples were collected by free percolation on two different dates, after harvest and after the Winter period.

The physicochemical parameters of water samples and of heavy metals in the rice grain were determined according to the analytical reference methods referred in Table 1.

3 Results and Discussion

3.1 Effect of Irrigation Water on the Quality of Soil Drainage Water

Physicochemical Parameters

The rice crop cycle was completed in 147 days. The mean values of pH, electrical conductivity (EC) and chlorides of the irrigation and drainage water of the pots (Tables 1 and 2) reveal significant differences between the treatments associated with the type of irrigation water. The significantly higher values of conductivity and chloride content of the water drained from pots irrigated with TWW are clearly associated with the origin of the irrigation water, which also has higher values than NW.

The pots irrigated with NW had a significant impact on the pH increase of the drainage water (from 6.7 to 7.2). In contrast, in the pots irrigated with same water (NW) by SDI conditions, the pH dropped from 6.7 to 6.1.

The total dissolved solids (TDS) were lower in the irrigation water compared to drainage, with the highest value in the TWW treatment. In turn, the total suspended solids (TSS) were higher in irrigation water, NW compared to TWW, possibly because TWW are filtered during the treatment process, with NW inversion of the order relationship in the drainage water (SDI-TWW> F-NW or SDI-NW) (Table 1).

There was an increase between the Electrical Conductivity (EC) values of TWW irrigation samples and the respective drainage, which was higher than those with NW. Regarding the NW, an increase was observed about EC values between the irrigation water and that of the respective drainage (1610μS/cm with the NW and 2100μS/cm with the TWW). Instead, the sodium adsorption ratio (SAR), being lower in irrigation water (NW and TWW) than in drainage water (SDI-TWW and F-NW+SDI-NW), and also increasing between irrigation water and the respective drainage, now has a higher increase in the case of NW (1.7) than in TWW (1.5) (Table 1). The salinity of the drainage water, deriving from that existing in the soil solution, increased due to the contribution of TWW high saline content in sodium and chlorides (sodium chloride), although other calcium and magnesium salts were also present (Table 1).

Table 1. Average values of the physical-chemical analysis of the water used for irrigation and the drainage water from the pots

Parameter (units)	Method	RV	MAV	Irrigation		Drainage	
				NW	TWW	SDI-TWW	(SDI,F)-NW
pH (Sorensen scale)	NP411:1966	6,5–8,4	4,5–9,0	6.7	7.2	7	6.6
Nitrates (mg/L)	ASTM D 4327.2017	50	—	2.9	30	66	10
Sulfates (mg/L)	ASTM D 4327.2017	575	—	20	54	230	130
Chlorides (mg/L)	ASTM D 4327.2017	70	—	43	150	730	470
Fluorides (mg/L)	ASTM D 4327.2017	1,0	15	<0.30	<0.30	<0.30	<0.30
Total Suspended Solids (TSS) (mg/L)	SMEWW 2540 D, 23ªEd	60	—	7.6	1.9	8	3.6
Salinity: Electrical Conductivity (µS/cm)	Ml n°013 (03.05.2011)	1	—	290	1100	3200	1900
Sodium Adsorption Ratio (SAR)	Calculation	8	—	1.5	8.5	10	3.2
Total Dissolved Solids (TDS) (mg/L)	Electrometry	—	—	150	650	1700	1000
Free residual chlorine (mg/L)	SMEWW 4500 G, 22.Ed	—	—	<0.1	<0.1	<0.1	<0.1
Aluminum (mg/L)	DIN EN ISO 11885	5	20	0.04	0.31	0.09	0.09
Arsenic (mg/L)	DIN EN ISO 11885	0, 1	10	<0.005	<0.005	<0.005	<0.005
Barium (mg/L)	DIN EN ISO 11885	1.0	—	0.042	0.055	0.15	0.15
Beryllium (mg/L)	DIN EN ISO 11885	0,5	1	<0.002	<0.002	<0.002	<0.002
Boron mg/L	DIN EN ISO 11885	0,3	3,75	<0.05	0.09	<0.05	<0.05

(*continued*)

Table 1. (*continued*)

Parameter (units)	Method	RV	MAV	Irrigation		Drainage	
				NW	TWW	SDI-TWW	(SDI,F)-NW
Cadmium (mg/L)	DIN EN ISO 11885	0,01	0,05	<0.001	<0.001	<0.001	<0.001
Calcium (mg/L)	DIN EN ISO 11885	—	—	12.7	52.1	102	133
Chromium (mg/L)	DIN EN ISO 11885	0,1	20	<0.005	<0.005	<0.005	<0.005
Cobalt (mg/L)	DIN EN ISO 11885	0,05	10	<0.005	<0.005	<0.005	<0.005
Copper (mg/L)	DIN EN ISO 11885	0,2	5	0.027	0.004	0.009	0.007
Iron (mg/L)	DIN EN ISO 11885	5.0	—	0.03	1.3	0.11	0.02
Lead (mg/L)	DIN EN ISO 11885	5	20	<0.005	<0.005	<0.005	<0.005
Lithium (mg/L)	DIN EN ISO 11885	2,5	5,8	0.01	<0.01	<0.01	<0.01
Magnesium (mg/L)	DIN EN ISO 11885	—	—	5.33	21.5	26.9	33.2
Manganese (mg/L)	DIN EN ISO 11885	0,2	10	<0.005	0.15	0.022	0.02
Molybdenum (mg/L)	DIN EN ISO 11885	0,005	0,05	0.009	0.009	0.009	0.009
Nickel (mg/L)	DIN EN ISO 11885	0,5	2	<0.005	<0.005	0.01	<0.005
Potassium (mg/L)	DIN EN ISO 11885	—	—	4.3	14.4	73.8	23.7
Selenium (mg/L)	DIN EN ISO 11885	0,02	0,05	<0.01	<0.01	<0.01	<0.01
Sodium (mg/L)	DIN EN ISO 11885	—	—	24.7	290	458	155
Vanadium (mg/L)	DIN EN ISO 11885	0,1	1,0	<0.005	<0.005	<0.005	<0.005
Zinc (mg/L)	DIN EN ISO 11885	2	10	0.16	<0.01	0.01	0.02

RV, Recommended Value; MAV, Maximum Allowable Value,
Irrigation water: NW, normal water; TWW, treated wastewater.
Drainage water: SDI-TWW, underground drip irrigation with TWW; (SDI,F)-NW, average values of underground drip irrigation and continuous flooding with normal water.

The SAR indicates the ratio of exchange sodium to other non-toxic cations, such as calcium and magnesium. As sodium degrades the soil structure and raises the pH, it increases the risk of immobilization and the consequent deficiency of some micro-elements, especially zinc.

The quality of drainage water with respect to EC and Chlorine (Table 2), improved after the Autumn/Winter period, without cultivation, during which the rainfall occurred allowed the leaching of the soil, with a very relevant reduction in salinity.

Table 2. Average values of physical-chemical parameters of irrigation and drainage water from the pots after harvest and at the end of the Autumn/Winter period.

Parameter	Irrigation		Drainage					
	2019*		At harvest/2019**			After Winter/2020***		
	NW	TWW	SDI-TWW	SDI-NW	F-NW	SDI-TWW	SDI-NW	F-NW
pH	6.70	7.20	6.60	6.10	7.20	5.97	5.74	5.92
ECondutivity(μS/cm)	290.0	1100.0	2758.0	1064.0	1128.0	202.2	278.0	324.6
Chlorine (mg/L)	43.0	150.0	541.0	165.0	184.0	19.2	41.0	56.4
Nitrates (mgNO_3/L)	<3.0	30.0	66.0	10.0	10.0	0.5	0.8	0.4
TDS (mg/L)	150.0	650.0	1000.0	1000.0	1700.0	145.6	195.6	229.4

Irrigation water: NW, normal water; TWW, treated wastewater;
Drainage water: SDI-TWW, underground drip irrigation with TWW; SDI-NW, underground drip irrigation with normal-water; F-NW, irrigation by continuous flooding with normal-water.
*samples of irrigation water, during irrigation season. Determinations made at the *Tomaz Laboratory*
**samples taken from 10/Oct to 10/Dec/2019. Determinations made at the ESAC Chemistry Laboratory
***sample taken at the end of the autumn/winter period. Determinations by the ESAC Chemistry Laboratory

The values of arsenic, cadmium, cobalt, lead, chromium, lithium, nickel, selenium and vanadium from TWW were below the detection threshold of the method, which leads us to conclude, in the first approach, that in relation to these parameters, this water has no restrictions for irrigation use. The only sample where boron was detected was in TWW (90 μg/L) used for irrigation, which was not in the drainage water of the pots irrigated with this water. It should be noted that the boron content of TWW did not exceed one part per million (1 ppm = 1 mg/L), a limit that should not be reached given its toxic nature for rice. The values of arsenic, chromium, lead found in irrigation and drainage water are below 0.005 mg/L, and those for cadmium below 0.001 mg/L. These values are below the maximum limits stipulated by the legislation [6, 7] (Table 3).

3.2 Effect of Irrigation Water Quality on Rice Production. Heavy Metals in the Grain

Total arsenic, cadmium, lead and mercury concentrations found in rice samples are shown in Table 4. Portuguese legislation sets for limits on the content of heavy metals in water for human consumption and irrigation water; however, limits for food products

Table 3. Maximum levels of heavy metals in the water, according to the legislation in force

Heavy metal	Irrigation		Potable for human consumption			Residual	Surface
	MRV (mg/L)	MAV (mg/L)	MRV (mg/L)	MAV (mg/L)	VP (mg/L)	VLE (mg/L)	MAV (mg/L)
Arsenic (As)	0.10	10	0.010	0.050	0.010	1	0.1
Cadmium (Cd)	0.01	0.05	0.001	0.005	0.005	1	0.01
Lead (Pb)	5	20		0.050	0.010	0.20	0.05
Mercury (Hg)	—	—	0.0005	0.001	0.001	0.05	0.001
Nickel (Ni)	0.5	2	—	—	0.020	2	0.05

VLE, emission limit value (concentration that must not be exceeded) DL-236/98
MAV, maximum allowable value (value that must not be exceeded) DL-236/98
MRV, maximum recommended value (value that must be respected or not exceeded) [6].
VP, parametric value (maximum residual concentration of the polymer in contact with water) [7].

are not yet defined. Considering the maximum levels of 0.2 mg/kg, fixed for inorganic arsenic, total cadmium and lead in rice by the Regulation of the European Communities [8, 9], it appears that lead is below this threshold in all samples.

Table 4. Concentration of total arsenic, cadmium, lead and mercury in the grain of brown rice produced in the three treatments and under traditional field conditions

Heavy metal (mg/kg)	SDI-TWW	SDI-NW	F-NW	FC
Arsenic	<0.1	<0.1	0.4	0.9
Cadmium	0.1	0.3	0.2	<0.01
Lead	0.1	<0.02	0.2	<0.02
Mercury	<0.00	<0.00	<0.00	<0.00

Treatments: SDI-TWW, underground drip irrigation with TWW; SDI-NW, underground drip irrigation with normal water; F-NW, irrigation by continuous flooding with normal water; FC, field conditions, produced by a rice farmer field, with flood irrigation under field conditions.

Mercury was not detected in any of the samples. In any case, there are no legislated maximum values for this metal for rice. The cadmium content of rice irrigated with NW (0.3 mg/kg) was above the limit for this heavy metal.

The soil of the SDI pots was kept in oxic conditions, which would have increased the solubility of this element, due to the favorable conditions for the desorption of aluminum, iron and manganese oxy-hydroxides or solubilization of CdS or $CdCO_3$. In this drained soil, it is procuced cadmium sulfate ($CdSO_4$), which, being soluble in water, allows its greater absorption by rice roots. Cadmium is strongly retained in flooded soils (reducing condition), accumulating less cadmium than that developed under oxidizing conditions, which is related to the formation of cadmium sulfide in anoxic conditions [10]. TWW had higher values of pH (7.2), aluminum (0.31 mg/L), iron (1.3 mg/L) and manganese (0.15 mg/L) compared to normal water, which had values pH 6.7, aluminum 0.04 mg/L, iron 0.03 mg/L and manganese less than 0.05 mg/L.

The arsenic content found in rice shows a direct relationship with growing conditions (aerobic or anaerobic). The rice produced in flooding contains the highest levels in pot (F-NW, 0.4 mg/kg) and in field conditions (0.9 mg/kg), well above the maximum limit of 0.2 mg/kg regulated for arsenic for rice [9]. Similar results had already been described by Simões [11] in traditional carolino rice (japonica variety) produced in Lower-Mondego Valley, Portugal, with average values of 0.2426 mg/kg.

The accumulation of arsenic in plants depends essentially on its bioavailability (arsenic speciation) and on the levels of arsenic present in the soil [12]. Given that in this trial the same soil (composition and type) was used in all treatments and that all irrigation water contained trace levels of arsenic (<0.005 mg/L) (Table 4), the differences found are due to the effect other factors that resulted in different arsenic bioavailability in treatments.

Under conditions of almost permanent flooding, in anaerobiosis, arsenic exists mainly dissolved in the form of arsenite, its most toxic and most bioavailable form for the plant [13], which justifies the capture of arsenic by cultivated rice in flooding with NW order of magnitude greater than that of the drip irrigated with TWW. Arsenic is carried by the phloem to the seeds where it is stored in vacuoles and other tissues of the edible parts of the grain. Their storage in the grain is done mainly in the form of arsenite and dimethylarsinic acid [14].

4 Final Considerations

The use of TWW in rice irrigation is a challenging issue. The experiment carried out in pots during the 2019 season, based on physicochemical analysis of the water samples, confirmed the soil salinity risk associated with TWW. The high EC of drainage water, with 2758μS/cm, when TWW was used to irrigate (corresponding 96μS/cm for NW), with a predominance of the elements sodium and chlorine. This issue is very important for productivity and soil conservation. It should be noted that after the autumn-winter period, in which the pots were subject to precipitation and free drainage, the EC values dropped to values similar to the initial ones. The results on heavy metals in irrigation and drainage water, lead to the conclusion that they are at levels below the maximum stipulated by legislation.

Regarding the levels of heavy metals detected in rice grains, it was found that the grain produced in the SDI-NW treatment has a cadmium content above the legal limit. The greater solubility of this element is explained by the redox potential and the pH of the soil in these test conditions, although no progress has been made in this evaluation. The arsenic level was higher in grains produced in the F treatment, whereas that value in the SDI treatment is insignificant. Results showed that the irrigation with TWW rises the EC of drainage water, but that the rice grain does not present increased risks to public health due to the low content of arsenic, cadmium, lead and mercury. However, the irrigation method must be adapted to a SDI to avoid human and animal contact with putative contaminants present in this water and thus safeguarding Environmental and Food Safety. The use of TWW must imply a careful plan to monitor the soil salinity throughout the irrigation campaign, to prevent critical situations of reduced production, or soil conservation. Long term soil effects of TWW, including winter percolation, will be assessed in the future.

Acknowledgments. Project MEDWATERICE: Towards a sustainable water use in Mediterranean rice-based agro-ecosystems. The project is financed in the context of the PRIMA Programme (PRIMA-Section-2018; Topic: 1.1.3: Irrigation technologies and practices). (MEDWATERICE-PRIMA-0005-2018, www.medwaterice.org).

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