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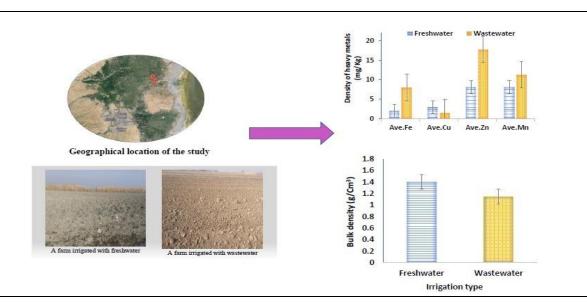
Original paper

A long-term study of the effects of wastewater on some chemical and physical properties of soil

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GRAPHICAL ABSTRACT



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ABSTRACT

Nowadays, reuse of wastewater is widespread to prevail over shortage of water and to fertilize agricultural lands. This study was conducted to investigate effects of wastewater on some chemical and physical properties of soil. For this purpose, two farms were selected. These farms are located in the Ghahremanloo region at Urmia plain, West Azerbaijan province located in northwestern of Iran. There is no exact information regarding total amount of wastewater delivered to these lands, but flooding irrigation employing wastewater was applied during growing season. The farms are irrigated with two treatments, including wastewater treatment and freshwater where the surface irrigation method was utilized to plant corn. Experiment design was conducted as completely randomized blocks. Each experiment was repeated four times for both freshwater and wastewater treatments. Results of this study showed that the use of wastewater results in a significant decrease in soil's electrical conductivity (EC), sodium absorption ratio (SAR), and a substantial increase in calcium carbonate equivalent (CCE) and organic materials (OM) of the soil. Besides, the wastewater decreased density of Cu and increased density of Zn, Mn, and Fe significantly, known as heavy metals of the soil. However, the density of these elements in the soil was below detection limit. Bulk density also showed a significant reduction in wastewater usage. Finally, paired t-test and Mann-Whitney nonparametric tests were implemented to validate data.

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1. Introduction

The use of wastewater for irrigation in agriculture in areas facing tension and water scarcity reduces the pressure on available water *Corresponding author Email: sara.habibi1987@yahoo.com resources. It allows high-quality water resources to be utilized for other uses and the development of security and health infrastructure. (Schacht and Marschner. 2015). Moreover, it has mentioned in several studies such as Nicholson et al. (2018) that that applying sewage

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sludge to agricultural lands is playing an essential role in nourishing the soil and maintaining soil organic matter levels without any adverse effects, in most of the cases, on crop quality. Therefore, due to recent droughts and the shortage and reduction of water quality in Iran, irrigation with refined sewage or so-called wastewater has become a common practice. However, it should be noted that although the use of wastewater for irrigation is a valuable strategy to raise water resources, the quality and conditions of this water can create challenges in agriculture. Irrigation with urban sewage can cause changes in soil properties that play an essential role in transferring, storage, and movement of nutrients in wastewater, in particular, the physical and chemical properties of soil, such as texture, structure, porosity, hydraulic conductivity, and mineral rate contained in it. The use of sewage in the soil can act as a suitable material and cause soil physical and chemical properties to be corrected. According to Urban et al. (2017) the utilization of the wastewater can lead to several advantages. First, wastewater has more nutrients than freshwater, and it can be considered as a source of nutrients for agriculture. Secondly, it can decrease the demand for chemical fertilizers as well as the release waste products into water bodies. Nowadays, in Iran, wastewater is used commonly and extensively in the irrigation of agricultural products and natural landscapes. Wastewater has different biological, physical, and chemical effects on the environment. The irrigation of the wastewater has certain benefits, such as the delivery of essential nutrients and organic matter, water and nutrient savings, and water pollution reduction (Khalid et al. 2018). It should be noted that using wastewater due to the existing contains toxic compounds such as zinc, lead, and mercury can result in serious health and environmental risks (Khalid et al. 2018). Even the modified wastewater is also different and worse than freshwater in salinity, alkalinity, PH, micronutrient density, foodstuffs, dissolved organic materials, and total suspended solids (Ben-Hur and Assouline. 2005; Halliwell et al. 2001). Since soil is a dynamic system of minerals, organic materials, water, air and microorganisms, wastewater, and sewage application, especially for a long time, change its physical, chemical, and biological properties. Considering the growing interest in using wastewater for irrigation and its potential effects on soil, various researchers have studied the impact of irrigation with wastewater on the physical and chemical properties of soils. Abegunrin et al. (2016) focused on assessing the physical and chemical properties of the soil as well as the growth parameters and water usage patterns in southwest Nigeria. The study was conducted regarding two indigenous vegetables (Eggplant and Spinach) being irrigated with three kinds of wastewater (abattoir wastewater, bathroom, laundry wastewater, and cassava effluent). Gharaibeh et al. (2016) investigated the changes in some properties of clayey soils such as aggregate stability, exchangeable sodium percentage, organic matter, electrical conductivity, hydraulic conductivity, and cumulative infiltration using treated wastewater and freshwater. While a significant decrease was observed in hydraulic conductivity and cumulative infiltration regarding the wastewater irrigation method, the other four mentioned properties showed a significant increase. There are other pieces of research in this context such as Klay et al. (2010), Hasan et al. (2014), and Aiello et al. (2007). Khodadai et al. (2015) investigated the effect of irrigation with urban and industrial wastewater (for eight years) and river water (twenty years) on some physical properties of soil in agricultural lands of Zarinshahr area of Lanjan. Irrigation with urban and industrial wastewaters increased bulk density, as well as, reducing the soil hydraulic conductivity, permeability, and buckling of soil moisture curves. Razzaghi et al. (2016) conducted their research investigating the long-term effect of wastewater on the physical and hydraulic properties of soil, comparing the influence of some infiltration models.

Peikert et al. (2017) investigated the degradation of the organic matter of olive mill wastewater as well as its phytotoxic and water repellent effects independence on four different climatic conditions with regard to the hypothesis that optimal biological activity is guaranteed through warm conditions with sufficient soil moisture leading to an optimum negative effect of the olive mill wastewater treatment. Hamada et al. (2018) studied the treatment efficiency of the Gaza wastewater treatment plant by considering influent input values of pH, temperature, biological oxygen demand, chemical oxygen demand, and total dissolved solids using Neural Network procedure. A study on evaluating the long-term effects of residual sewage sludge was conducted by Florentino et al. (2019), taking both chemical attributes and potentially toxic elements into account. Bozkurt and Cimrin (2003) focused on nutrients and heavy metals of calcareous soil such as Mn, Zn, Cu, Ni, Cd, and Cr. They investigated these materials being affected by sewage sludge and barnyard manure in the ten-year-old Starking

Delicious orchard located in Turkey. Urbano et al. (2017) assessed the changes in physical, chemical, and microbiological characteristics of a Dusky Red Latosol, the yield, and the quality of lettuce after cultivation with treated wastewater on irrigation. They considered parameters such as odium adsorption ratio, magnesium, Ca, Na, K, nitrate, chlorate, PH, EC, total coliforms, Escherichia coli, chemical, and biochemical oxygen demand were analyzed in irrigation water. Aydin et al. (2015) evaluated the heavy metal accumulation, including Pb, Cd, Cr, Cu, Ni, Zn, Hg, where the considered nine sites had been irrigated with wastewater for 40 years. In a more recent study, Sinegani et al. (2018), explored the KNO3 and DTPA extractable heavy metals and compared the results in a saline soil sampled to a non-saline agricultural soil. Todeshki et al. (2015) considered the effect of treated and magnetic wastewater on the soil properties, through randomized complete block design with three irrigation treatments as wastewater, magnetic wastewater, and normal water in four replications. They observed no statistically significant difference between the treatments regarding the amount of soil salinity, but the subsurface layers were imposed on a substantial change of salinity in the magnetized wastewater treatment. Previous research studies have shown that the use of refined sewage is prevalent throughout the world, but what matters is its solutions, how it is used, and its localization based on the products used and the physical and chemical conditions of the soil.

Currently, in many cities of Iran, urban wastewater and surface runoffs that leave the cities are used in agricultural fields. In this regard, due to the above notes and with consideration of this fact that in developing countries in addition to treated wastewater, raw wastewater is also increasingly used in agriculture, this research has been done for further familiarity to raw wastewater effect on some physical and chemical properties of soil including PH, SAR, bulk density of soil, soil tissue, organic materials of soil, equivalent calcium carbonate, electrical conduction, and density of low consumption metals in the soil profile.

2. Materials and methods

In order to study the consequences of irrigation with wastewater on the chemical and physical properties of the soils of the Ghahramanloo region, two pieces of land with a distance of 650 meters from each other were chosen. Olya Ghahramanloo is a village in environs of the central part of Urmia city of West Azerbaijan province of Iran. One of the farms, for ten years, was under the impression of irrigation with raw wastewater from the wastewater channel near the farm, and the other was irrigated with freshwater. Both farms have the same products and irrigation methods. The plan area has semi-dry weather and plain lands, an average rainfall of 330 mm/year. The average annual temperature in the region is 11.8 °C.

The land of the first piece had been irrigated with raw wastewater for ten years, but for the other land, no kind of irrigation with wastewater has been used. In both plan pieces, the experiment was done for four repetitions that the freshwater and wastewater were chosen as witness and treatment, respectively, and the index plant in this plan was corn implanted by hand. In each area of study, for measurement of desired properties, some manipulated and un-manipulated samples were prepared. The samples were prepared from the depth of (10-30) centimeters in four repetitions to define the tissue of both pieces. The texture of the soil was determined by the hydrometric method. In Table 1, the texture and percentage of primary particles in the soils of the farms are presented. The resulting samples of water treatment that were used for irrigation were transferred to the laboratory and tested. The un-manipulated sample has measured bulk density. After drying for 24 hours (at 105 °C temperature), the bulk density was defined by the following formula.

$$B_{d} = M_{s}/V_{T}$$
(1)

where, $M_{\rm s}$ is soil mass, and $V_{\rm T}$ is the total volume of the cylinder. Chemical properties of soil in chosen situations concluding PH, EC, and CaCO₃ were measured by standard methods of Page (1982). For statistical analysis of obtained results in each farm, irrigated by freshwater and wastewater, a statistical software called Minitab 16 was used. A comparison test was done by the t-test and nonparametric Mann-Whitney at a confidence level of 0.05 and 0.1.

2.1. Statistical tests

In using and choosing statistical tests, one of the fundamental and principal assumptions of parametric methods is the normality of data or, in other words, before each test, which is done by assuming that data is normalized, a normal test should be accomplished. This assumption is usually considered before test selection. If data are not normal, equivalent nonparametric methods must be used. For this purpose, in this paper, the Kolmogorov-Smirnov test was applied. Kolmogorov-Smirnov test with the Chi-squared test is a well-fitting test. However, considering to Chi-squared test limitations, usually for normality test, Kolmogorov-Smirnov is used. Kolmogorov-Smirnov test shows the normality of the distribution of data. This means it compares the distribution of an adjective in a sample (here the amount of material in wastewater sample or freshwater sample) with the distribution, which is assumed for the society (for example, the amount of EC in wastewater and freshwater). If data has a normal distribution, the parametric test is applicable; otherwise, the nonparametric test should be used. For normality test, statistical assumptions are adjusted as follows: H₀: data distribution of each variable is normal.

H1: data distribution of each variable is not normal.

Table 1. Comparison of the particle size distribution and soil texture class irrigated with freshwater and wastewater

Region	Iteration	Sand	Clay	Silt	Soil texture class
Irrigotod	1	47.5	20	32.5	
Irrigated with	2	50	20	30	Loomy
freshwater	3	45	22.5	32.5	Loamy
	4	47.5	22.5	30	
Irrigated with wastewater	1	30	30	40	
	2	35	32.5	32.5	Clay loamy
	3	32.5	32.5	35	Ciay IOanty
	4	32.5	35	32.5	

By performing this test, the software calculates a meaningful number. In the normality test, if meaningfulness is more than confidence level (for example, 5 % or 0.05), the normality of distribution is received. Nonparametric tests are kinds of tests in which data have abnormal distribution and are less identifier than parametric tests (like Mann-Whitney and Kruskal-Wallis tests). The t-student test is performed for evaluation of the sameness and goodness of fit for the average sample with average society when the standard deviation of the society is unknown. In order to apply this test, the study variable should be in the interval scale, with normal distribution and the number of the sample less than 30. The Mann-Whitney test is used to compare differences between two independent groups when the dependent variable is either ordinal or continuous, but not normally distributed. For example, the Mann-Whitney test can be applied to understand whether attitudes toward pay discrimination, where attitudes are measured on an ordinal scale, differ based on gender (i.e., your dependent variable would be "attitudes towards pay discrimination," and your independent variable would be "gender," which has two groups: "male" and "female").

The Mann-Whitney test is often considered the nonparametric alternative to the independent t-test, although this is not always the case. In order to know about normality or abnormality of data by Minitab 18 software, normality graph is drawn, and by obtained p-value, we can find their conditions. We use a t-test if both data are normal and the Mann-Whitney test provided that both or one of the data are normal to identify whether there is a significant difference between the two societies or not. Both of these processes are performed by Minitab 18 software, which the results are shown in Table 2. For example, consider the EC parameter. As is seen in Fig. 1, the p-value is more than the confidence level (including 0.05 and 0.1); therefore, in both of these levels for the function, we can consider a normal distribution. The p-value for wastewater, Fig.2, is also more than the confidence level (including 0.05 and 0.1).

 Table 2. T-test to test the mean equality of societies in a confidence level of 95 %.

Paired T-test and CI: Freshwater value, Wastewater value				
Paired T for Freshwater value – Wastewater value				
Ν	Mean	StDev	SE Mean	
4	3.375	0.250	0.125	
4	1.770	0.185	0.093	
4	1.605	0.251	0.125	
95 % CI for mean difference: (1.206, 2.004)				
	iter val N 4 4 4 4	ter value – Was N Mean 4 3.375 4 1.770 4 1.605	N Mean StDev 4 3.375 0.250 4 1.770 0.185 4 1.605 0.251	

T-test of mean difference= 0 (vs not zero): T-value=12.81 P-value= 0.001

As it is seen according to the given explanations above and output values of the test, which in both cases is less than 0.05 and 0.1 $\,$

respectively, zero assumption is rejected in confidence levels, and it is concluded that there is a significant difference between averages of the two societies.

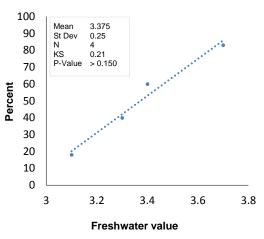


Fig. 1. Testing the normality of EC on the land irrigated with freshwater.

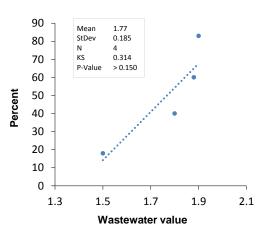


Fig. 2. Testing the normality of EC on the land irrigated with wastewater.

3. Results and discussion

In the first section of this investigation chemical properties of the study lands including pH, electrical conductivity (EC), equivalent calcium carbonate (ECC) and some metals have been explored, and in the second part physical properties of soil including texture, organic materials and bulk density (BD) of soil are investigated.

3.1. Wastewater effect on chemical properties of soil

The results of experiments show that irrigation with wastewater changes almost all of the chemical properties of soil, while irrigation with freshwater does not. Some of the chemical properties of water treatment like pH, EC, and SAR, and their quality evaluation are seen in Table 3. pH is a simple parameter for determining the quality of irrigation water, which shows acidity and alkalinity. The normal range of PH for irrigation water is between 6.5 -8.5. Out of this range, the warning signal indicates the bad quality of water. To study wastewater and freshwater treatment, guidelines for the evaluation of irrigation water, WHO is used. Every three parameters, pH, EC, and SAR are in acceptable ranges based on WHO standards.

Table 3. The main chemical composition of raw wastewater and	
freshwater used for irrigation.	

neonwater doed for inigation.					
Parameter	Freshwater	Wastewater	WHO		
pН	7.12	7.5	7.6		
EC, dS/m	2.84	1.55	3		
SAR	1.25	0.72	>3		

3.2. The density of low consumption metals in the soil profile

Wastewater irrigation effect on the accumulation of heavy metals in soil depends on different factors like the density of heavy metals in wastewater, length, irrigation duration, the texture of pH, and cation exchange capacity of the soil. In fact, 10 to 50 years are needed until the heavy metals level of irrigated soil with wastewater exceeds the permissible limit. For example, Rattan et al. (2005) results showed that irrigation with wastewater increases iron density only in the soil after five years, density of heavy metals like zinc, iron, nickel, and lead after ten years and density of heavy minerals like iron, zinc, copper, manganese, nickel, and lead after 20 years. The density of metals in soil and statistical analysis of changes in each land irrigated by freshwater and wastewater have been shown in Table 4. By examining Table 4, we can conclude that copper density in the area irrigated by wastewater is less than by freshwater, and this conclusion can be related to the higher amount of this metal in freshwater. While using wastewater had a more significant effect on the amounts of minerals like iron, manganese, and zinc than freshwater. The interaction of wastewater and freshwater treatments application for irrigating the plan farms on soil metals have been shown in Fig. 3. In this figure, we can observe that the average comparison test has shown a significant difference between using wastewater and freshwater treatments, with more iron, manganese and zinc, and less copper in wastewater. Statistical analysis of the t-test showed that there is a significant difference of 5 % between treatments' effects on the density of metals in soil.

3.3. pH

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As stated in Kunhikrishnan et al. (2012), the impact of wastewater irrigation on soil pH depends on the PH of the wastewater source and the pH buffering capacity of the soil. While in some papers such as Bedbabis et al. (2015), the pH increases by applying a wastewater treatment, there are other investigations that report no changes in the pH of the soil such as Bedbabis et al. (2014) and Urbano et al. (2017). Following the latter research, no significant difference in soil pH was observed unless in two points between the irrigated region by

freshwater and wastewater. This can be due to the reason that wastewater and freshwater pH are close to each other. Table 5 shows the average value of soil pH for the two treatments. As it is seen, the average amount of pH in plan farms soils does not have a significant difference. This conclusion can be due to this reason that there is not any significant difference between freshwater and consumed wastewater pH.

3.4. Electrical conductivity

A significant increase in the EC value of soil was observed in a land irrigated by freshwater (Table 5). This finding is in contrast with those reported by several studies (Bedbabis et al. (2015) and Raeisi-Vanani et al. (2017)) As it is shown in Table 5, freshwater used for irrigation has more EC value than wastewater; therefore, the EC increase in the land irrigated by freshwater is not surprising. The salinity conditions of soils of plan farms by applying treatments. Moreover, soil salinity analyzing results show that in a farm irrigated by freshwater, the soil is saltier. Also, a significant difference is observed between farm soils of two freshwater and wastewater treatments based on a statistical-paired t-test at 5 % level.

3.5. Equivalent calcium carbonate (CCE)

Calcium carbonate has been significantly increased more by using wastewater irrigation than freshwater (Table 5). Accumulation of calcium carbonate in dry and semi-dry regions is related to some complicated processes depending on soil properties, maternal materials, rainfall, and fluctuations in groundwater. The accumulation of calcium carbonate in the region irrigated by wastewater may depend on decomposition processes. In the area, maternal calcareous materials are transferred to a specific depth of topsoil due to plowing and therefore increasing calcium carbonate in the land irrigated by wastewater. These results are compatible with Bedbabis et al. (2015) and Raeisi-Vanani et al. (2017) studies where wastewater treatment increases the CaCO₃ of the soil.

Region	Iteration	Fe, mg/kg	Cu, mg/kg	Zn, mg/kg	Mn, mg/kg
	1	1.38	2.42	6.36	8.78
Irrighted with freehunder	2	2.66	3.26	8.76	6.2
Irrigated with freshwater	3	1.96	2.98	7.92	7.76
	4	2.04	3.22	9.38	9.6
Average		2.01	2.97	8.105	8.085
Standard deviation		0.523	0.386	1.308	1.464
	1	7.24	1.96	21.6	12.3
Irrigated with wastewater	2	7.6	1.36	15.96	10.3
ingaled with wastewater	3	9.02	1.14	18.3	10.54
	4	8.2	1.56	14.72	11.94
Average		8.015	1.505	17.645	11.27
Standard deviation		0.778	0.348	3.025	0.997
EPA		5-50	10-80	10-300	200-10000

Table 5. Changes in soil's OM, PH, EC, and CCE in freshwater and wastewater treatment.	
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Region	Iteration	OM, g/Kg	PH	EC, dS/m	CCE, %
	1	33.152	7.1	3.1	12.5
Irrigated with freshwater	2	35.919	7.12	3.4	12
inigated with neshwater	3	39.272	7.3	3.3	10.5
	4	37.771	7.1	3.7	11.5
Average		36.528	7.155	3.375	11.625
Standard deviation		2.635	0.097	0.25	0.853
	1	47.31	7.3	1.8	32.5
Irrigated with wastewater	2	45.962	7.1	1.9	30
ingated with wastewater	3	46.84	7.6	1.5	32
	4	48.424	7.7	1.88	32.5
Average		47.149	7.425	1.77	31.75
Standard deviation		1.029	0.275	0.185	1.190

3.6. Organic materials of soil

Organic materials in the farm irrigated by wastewater are about 1.2-1.4 times more than in a farm irrigated by freshwater (Table 5). This outcome is in line with the result obtained by Bedbabis et al. (2015), In this field, the changes in abundance of clay, type of clay minerals, soil pore size distribution, and type of organic material can be useful factors in decomposition and supply of organic materials. Others have observed similar results considering the changes in the OM portion in the soil. Yang et al. (2015) reported a 7.1 % increase of OM in the soil irrigated with sewage compared to agricultural soil that irrigated with groundwater. The more growth in the number of organic materials in a land irrigated by wastewater can be related directly to the high amount of organic materials in wastewater used for irrigating and also indirectly to organic materials of dense vegetation in the irrigated farm with wastewater. Based on Table 5, the number of organic materials in farm soil irrigated with wastewater is more than farm soil soaked with freshwater. This shows that the OM percentage of the wastewater to some extent affected the increase of OM percentage in soil irrigated with this treatment, and the continuity of wastewater usage in soils of the region is also useful in the renovation of organic materials. Other similar results can be obtained from Rattan et al. (2005) and Xu et al. (2010).

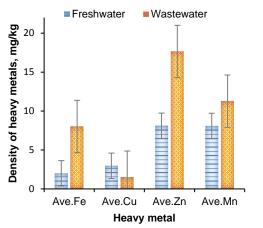


Fig. 3. Comparison of the density of heavy metals in soils irrigated with wastewater and freshwater.

3.7. Soil's texture

Since soil's behavior, to some extent, depends on particle size, this is so important to know the type of soil particles and their relative sizes. Therefore, the kind of soil tissue of the two regions was investigated separately. The soil tissues in both areas were defined, which the related information is shown in Table 1. The region irrigated by freshwater has loamy tissue, but the region irrigated by wastewater has clay loam tissue soils.

3.8. The bulk density of soil

The bulk density of soil (BD) in the farm irrigated with wastewater was significantly less than the bulk density of soil in the farm irrigated with freshwater (Table 6). Other researchers have obtained similar results. Mays et al. (1973) reported that by adding wastewater to the soil, the BD of soil was decreased. They attributed this decrease of the BD of soil to the formation of soil aggregates. In a more recent study, Mirzaei-Takhtgahi et al. (2018) showed that saturated hydraulic conductivity ratio would be increased using sewage, but the bulk density will be decreased significantly. Results obtained from the comparison test of BD averages by applying two treatments (Fig. 4) show that wastewater usage in soil decreases this parameter. The results are also in line with Andrew et al. (2016).

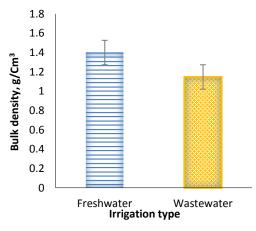


Fig. 4. Comparison of the mean bulk density in soils irrigated with wastewater and freshwater.

Table 6. The effects of irrigation on soil bulk density.				
Region	Iteration	BD, g/cm ³		
	1	1.42		
Irrigated with freshwater	2	1.37		
ingaled with reshwater	3	1.34		
	4	1.47		
Mean		1.4		
Standard deviation		0.057		
	1	1.11		
Irrigated with wastewater	2	1.18		
inigated with wastewater	3	1.14		
	4	1.16		
Mean		1.147		
Standard deviation		0.029		

4. Conclusions

In this research, irrigating with wastewater in comparison with freshwater, improved almost all of the physical and chemical properties of soil. Following by irrigating with wastewater and therefore adding more organic materials to the soil, the bulk density of soil was significantly decreased. Heavy metals density like iron, manganese, and zinc in a land irrigated with wastewater is considerably more than in an area irrigated by freshwater. However, existing metals in both properties are below the EPA standards. The soil irrigated with wastewater has less electrical conductivity and sodium absorption ratio than freshwater. Wastewater treatment not only does not produce a particular problem on salinity and alkalinity in soils of the region but also it decreases the soil salinity. Therefore, considering the dual nature of wastewater i.e. its positive role as a water supply source and its negative role as a pollutant, its application in irrigating should be performed based on a proper management, physical-chemical and even microbial adoption of wastewater with international standards and finally by taking into consideration that any region has a specific kind of water, soil and plant.

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