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Effects of a 25-year application of treated wastewater on soil properties of Cebala-Borj Touil irrigated perimeter (North Tunisia)

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ABSTRACT

In arid and semi-arid areas, the reuse of treated wastewater (TWW) is one of the solutions to the problem of water scarcity. Nevertheless, this resource may represent some risks to the environment and human health. In Tunisia, the use of TWW for irrigation has started since the sixties but its impact on soil quality is still not well understood. This study aims to investigate the effects of long-term irrigation with TWW on soil properties of Cebala-Borj Touil irrigated perimeter (North Tunisia). The studied soil is a fluvisol irrigated for 25 years with TWW. Water used for irrigation presents a high organic load, an alkaline pH and a relatively high salinity (3.73 mS/cm) but with trace metals concentration which does not exceed Tunisian and FAO standards. Two profiles were digged: P1 represents the soil irrigated with TWW and P2 is the control (non-irrigated soil). Soil samples were taken from each horizon and analyzed for their granulometric composition, pH, electrical conductivity (EC), trace metal elements (TME) and total organic matter (OM) content. Soil color and structure were equally described. The comparison of the two profiles showed that the irrigated soil presented a lighter color, a different structure and a slightly finer texture, mainly in surface layers, than the control. Besides, an acidification of soil surface and a problem of salinization/sodication accompanied with an OM accumulation in soil deep horizons were observed. TME concentrations were under the detection limit except for Mn, which showed an increase in the top horizon of P1 as compared to P2. Results show that the soil properties were seriously affected after 25-years irrigation with TWW, which must be improved by some management measures.

Keywords: Semi-arid region; Treated wastewater; Irrigation; Soil properties; Organic matter

1. Introduction

Population growth and economic development exert unprecedented pressure on water resources particularly in arid and semi-arid regions. Fresh water is becoming, therefore, a limited commodity in these areas, moreover water scarcity will be a striking incident, especially in the context of global warming. Hence, it is necessary to find alternatives to satisfy the strong water demand [1]. The reuse of treated wastewater (TWW) can be one of the solutions, often less expensive than the mobilization of a new resource [2]. The application of TWW has known a remarkable development in recent years and has been considered as a common procedure in many countries throughout the world. Indeed, the use of this non-conventional water was a very attractive practice and can be suitable for a large variety of applications (residential uses, groundwater recharge, aquaculture industrial cooling water, drinking water production, etc) [3]. Among the most common reuse applications is irrigation, which has been largely applied to agriculture due to the advantages related to nutrient recovery possibilities, socioeconomic implication, reduction of chemical fertilizer application, and effluent disposal [4]. Besides these benefi-

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cial effects, TWW have many disadvantages and are often associated with environmental pollution problems and health risks [5]. Some of these problems include eutrophication, which can stimulate the growth of algae, temporary oxygen deficits and unwanted changes in aquatic populations. The main health risks are associated with the contamination of crops or ground waters by the undesirable chemical constituents and pathogens, which can be found in TWW [6].

The application of TWW to irrigate agricultural soils is not a new practice [2]. In Tunisia for instance, the reuse of TWW in irrigation has been used since the 60s to irrigate lemon crops in the Soukra area [7]. In 1989, Tunisia was the first country in the southern banks of the Mediterranean to adopt regulations for reuse of TWW. Currently, irrigation with TWW is one of the methods, which are widely used as reliable water source [2]. It can increase the available water supply or release better quality supplies for alternative uses. Nevertheless, the main concern with the reuse of TWW in irrigation is its possible effect on the soil [8]. Indeed, this water may contain high levels of salts, toxic ions and organic residues [9]. Accumulation of these pollutants in soil can pose a threat to agricultural production. However, the pollutants content of TWW used for irrigation is not the only parameter of which depends the soil pollution. The contaminants content in soil depends also on soil composition, on physicochemical parameters and on the mechanisms that govern their distribution and their transport [10]. Moreover, the long use will serve as another parameter of soil pollution. In this regard, several studies have been conducted to assess the impact of long-term irrigation with TWW on the environment, and soil notably, which has been much less studied [11–13]. A number of risk factors have been identified in reuse of TWW for irrigation [14]; some of them are short term impacts (e.g., microbial pathogens) whereas others have longer-term impacts that increase with the continued use of TWW (e.g., salinity effects on soil, degradation of soil structure) [15]. Indeed, over time TWW irrigation can affect some soil properties and influence soil quality. It leads to a decrease in soil pH in alkaline soils and to an increase in acidic soils [16]. In fact, the soil pH increase as a result of numerous years of TWW application can be attributed to the high content of alkaline cations in TWW such as Na^+, Ca^{2+} and Mg^{2+} [17]. Carbonate ions combined with calcium or magnesium will precipitate as calcium carbonate ($CaCO_3$) or magnesium carbonate ($MgCO_3$). This will cause an alkalizing effect and will slightly increase the pH level [17]. However, soil acidification is due to the leaching of limestone by the leaching water. TWW can supply ammonium anion to soil, which is another source of soil acidification [18]. The decrease of soil pH is also explained by the low C/N ratio of effluents and the subsequent enhancement of the organic mineralization substances [4]. Thus, TWW adds usually large amounts of major and micro-nutrients to the soil, which stimulate the microbial activity and promotes soil OM mineralization [19]. This may cause the decrease of the soil cation exchange capacity [20]. Infiltration rate and hydraulic conductivity may also decrease through physical blocking of soil pores as a result of high loads of suspended solids (colloid clay and algal cell particles) during soil application of TWW [1,21]. Moreover, the higher levels of salt in TWW can increase soil salinity and sodicity, leading to reduced agricultural productivity and to degrade soil structure. In fact, clay dispersion is very sensitive to low level of sodicity and by that the susceptibility of aggregates to slaking [10]. By the same way, TWW usually contains high concentration in TME [22]. These elements can accumulate in the topsoil horizons in such manner that they reach their critical level for plant growth. The ETM mobility, adsorption, precipitation and leaching depends on the variability of soil structure, soil texture, soil composition and the physicochemical process. For example, the high ETM concentrations in soil are usually related to the increase of percentage of the OM content, which play an important role on ETM immobilization [23]. Further, the soluble organic constituents are going to favor the ETM mobility and to increase their concentration in solution [23].

Such is the framework where our research aims to evaluate the impact of long-term application of TWW on soil properties of Cebala-Borj Touil irrigated perimeter localized in North Tunisia. We believe a novel element in this study is the investigation of long-term effect of the TWW at the scale of the soil profile, while in a majority of researches, only the surface horizon was considered. This work was conducted in two distinct but complementary approaches: the first approach was assessing the quality of TWW for the irrigation of the perimeter, and the second was oriented to the study of the impact of these waters on soil properties.

2. Materials and methods

2.1. Study area

Our study site is the perimeter of Cebala-Borj Touil, located in the North of Tunisia (Ariana Governorate) (Fig. 1). It covers an area of 2800 ha. It is bordered in the north by Oued Medjerda, in the south by the reliefs of Nahli, in the east by a depression called Garat Ben Ammar and in the west by the GP8 road. The region has a semi-arid climate (mild winter, hot summer) with monthly air temperature ranging from 17°C to 20°C, dry summer and annual rainfalls around 488 mm mostly occurring from December to February. The average annual potential evaporation of 1306 mm, combined with the low rainfall and high temperatures makes irrigation essential for crop production. The soil is a fluvisol developed on calcareous-silty-clay recent alluvions, deposited by Oued Medjerda River.

The selected study area produces alternate cycles of crops, with successive winter and summer harvest of annual crops (oat, sorghum) sectioned every 10 years by a 3-year-long cropping of alfalfa. The water used for irrigation of the perimeter is a TWW originating from two treatment plants: Charguia, and Chotrana, localized near the studied area. Irrigation is applied during the dry season, by flooding through furrows. The mean annual application doses vary between 2000 and 3000 m^3 depending on crop. The study area was irrigated since 1990 and in order to assess the effects of the TWW, a nearby soil is taken as a control, which produces only olives and has been preserved from any source of irrigation.

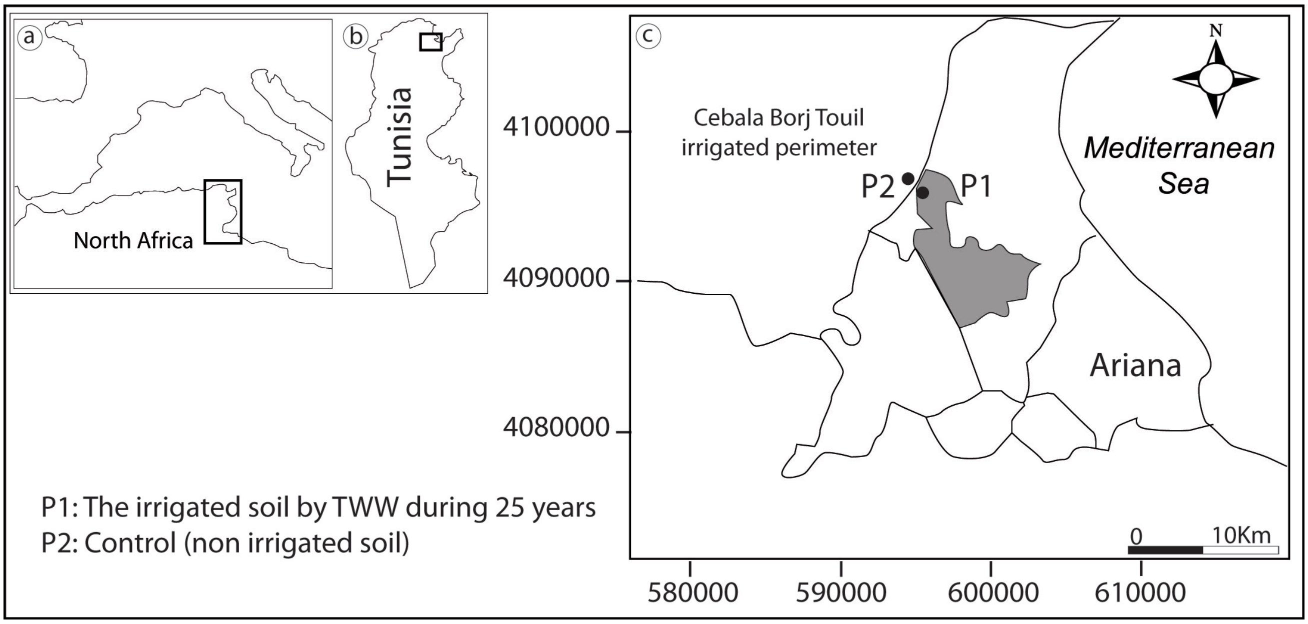

Fig . 1. (a) Map location in the Mediterranean basin, (b) Geographic situation of the study area, (c) Position of the studied soil profiles.

2.2. Water sampling and chemical analysis

TWW samples were collected from several valves of the farmer. In order to study the change occurring during our experimental period (2014–2015), TWW was sampled over time in May (2014), August (2014) and January (2015).

Temperature and dissolved oxygen content were measured in situ. Samples were kept at 4°C for the chemical analysis according to international norms. We did three replications of each analysis. pH (NF T90–008) and EC (NF EN 27–888) were measured by Electrochemistry method. Cations (Mg^{2+}, Ca^{2+}, Na^{+}, K^{+}) and anions (Cl^{-}, SO_4^{2-}) were measured by ionic chromatography (NF EN ISO 14911). Alkalinity attributed to bicarbonate (HCO_3^-) was estimated by Titrimetry method (NF EN ISO 9963-1). The precision of chemical analysis was checked by calculating the ionic balance, accepting error lower than 5%. The Biological Oxygen Demand (BOD_5) was measured by electrochemistry method (NF EN 1899-2), the Chemical Oxygen Demand (COD) was measured by titrimetry (NF T90-101) and Total Suspended Solids (TSS) were measured by the filtration device (NF EN 872). The Trace Metal Elements (TME) (Mn: manganese, Cr: chromium, Pb: lead, Zn: zinc, Ni: nickel, Cu: copper, Fe: iron, Cd: cadmium, Co: cobalt) were determined by atomic absorption after aqua regia acid digestion (NF EN ISO 15587-1).

2.3. Soil sampling and physico-chemical analysis

In this study, two profiles were explored: the first profile (P1) is the irrigated soil by TWW during 25 years. The second profile (P2) was considered as a control. For both sites, soil samples were taken from different horizons of the profile. Then, soil samples were air dried, sieved through a 2 mm diameter mesh and stored in sealed containers at 5°C. Sub samples of the soil fine fractions (< 2 mm) were stored for characterization. The following analytical methods were used according to international norms: Total Nitrogen (TN) was determined by the modified Kjeldahl method (NF EN ISO 11261). The Total Organic Carbon (TOC) was estimated by the sulfochromic oxidation method (NF EN ISO 14235). The soil texture was determined using the Robinson pipette method after oxidation of the Organic Matter with H_2O_2 and clay dispersion by hexametaphosphate solution (NF X31-107). Texture was expressed as percentage of sand, silt and clay. TME were measured by atomic absorption spectrometry method (NF ISO 11047). The soils were characterized for lime ($CaCO_3$) contents with the volumetric method (NF EN ISO 10693), electrical conductivity (EC) (NF EN ISO 11265) and pH (NF EN ISO 10390). We did three replications of each analysis. Besides, soils were characterized for their structure and their color using Munsell soil color charts.

3. Results and discussion

3.1. Water characteristics

To give a precise idea about the TWW quality used for the irrigation of Cebala Borj Touil perimeter, we made a statistical survey of geochemical parameters analyzed by the Regional Office of Agricultural Development of Ariana governorate for 10 years (2005–2015) [24]. According to the results and statistical study, the TWW composition was slightly the same during the analyzed period (2014–2015). The geochemical variables of the TWW used for irrigation are given in Table 1. TWW was, on average, alkaline with a basic pH value of 7.8 (±0.2). It was within the acceptable interval for irrigation, which range from 6.5 to 8.4 according to the FAO guidelines. The applied TWW presented a Na–Cl hydrochemical facies. Salinity, measured as EC, appeared relatively high (3.73 mS/cm) which may present a risk of soil salinization [10]. Nevertheless, the calculation of the Sodium Adsorption Ratio (SAR), shows a value of this ratio

Table 1
Geochemical variables of the TWW used for irrigation

	TWW	NT 106.03 norms
pH	7.8	6.5–8.5
EC (mS/cm)	3.7	7
SAR	6.7	–
BOD_5 (mg/L)	75	30
CDO (mg/L)	133.4	90
TSS (mg/L)	90	30
Mg^{2+} (meq/L)	7.1	–
Ca^{2+} (meq/L)	5.3	–
Na^{+} (meq/L)	17.2	–
K^{+} (meq/L)	1.0	–
SO_4^{2-} (meq/L)	10.5	–
Cl^{-} (meq/L)	14.1	–
HCO_3^{-} (meq/L)	6.2	–

EC: electrical conductivity, SAR: sodium adsorption ratio, BOD_5: biological oxygen demand during 5 d, COD: chemical oxygen demand, TSS: total suspended solids, Mg^{2+}: magnesium, Ca^{2+}: calcium, Na^{+}: sodium, K^{+}: potassium, SO_4^{2-}: sulfate, Cl^{-}: chloride, HCO_3^{-}: bicarbonate

inferior to 10. It was under the limit recommended by the European Commission for irrigation water (6.0–12.0), which indicates irrigation with this water constitutes a feeble risk of soil sodication. The concentrations of TME in TWW, as well as in the TSS were under the limit recommended by the Tunisian and the FAO standards. The values of BOD_5 (75 mg/L) and COD (133.4 mg/L) of the water sample are very high and well above the limits fixed by the NT 106.03 (Tunisian norms for TWW reuse in agriculture) at 30 mg/L and 90 mg/L respectively for BOD_5 and COD. Such a high organic load in water may cause an increase of the microbial biomass which may cause a reduction of the hydraulic conductivity and infiltration capacity, and therefore, cause physical and/or biologic clogging phenomena in soil [25]. The value of TSS (90 mg/L) also exceeds the Tunisian standard for the reuse of TWW. Indeed, the high content of TSS in water can clog irrigation infrastructure, especially if sprinklers or drip irrigation is used. In addition, if these solid materials are not biodegradable, they accumulate on the soil surface (formation of biological crust), which may lead to the clogging of the pores of the surface layers, and thus, they can decrease the percolation [21].

3.2. Soil characteristics

The comparison of the two studied profiles showed that the irrigated soil (P1) presented a lighter color on the surface and darker in the deep horizons than the control (P2) (see section on OM below). The P1 has an angular to sub angular structure in the surface, which became massive in the bottom while P2 maintains a granular structure.

Table 2 shows TME concentrations of the studied soils. TME (Cr, Pb, Zn, Ni, Cu, Fe, Cd and Co) values were not consistently affected according to the length of the period of the TWW application. They were usually under Tunisian standards but the Mn content showed a significant increase (29.3 mg/l) in the top horizon of P1 compared to P2 (23.6 mg/l). In general, heavy metals combine with other anions to form stable substances and tend to accumulate in surface layers [26]. Furthermore, their uptake by plants increases with decreasing pH (see section on pH below), due to the dissolution of metal-carbonate complexes, that releases free metal ions into the soil solution [22]. Indeed, ETM brought by TWW to the soil are not all in an available or bioavailable form. Their chemical form depends on the intrinsic soil conditions such as pH, OM content and the rate of clay [27].

Table 3 shows the main physico-chemical properties of the studied soil profiles. In P1, soil texture was slightly finer, mainly in topsoil compared to P2. Indeed, [1] stated that long-term application of TWW would produce more fine particles mostly in surface horizons. This was due to the change in pore size distribution as a result of the expansion and the dispersion of soil aggregates. The authors added that the collapse of soil surface structure was due to the OM dissolution.

Besides, the comparison of the two soil profiles reveals no effect of irrigation with TWW on almost all soil proprieties except pH, salinity and OM content.

3.3. Effect of irrigation by TWW on soil pH

The pH of the two profiles was slightly basic with values that became less important in the surface horizons of P1 and more significant in depth horizons compared to P2 (Table 3). Indeed, the irrigation by TWW caused mild acidification at the surface by lowering the pH. These findings are in agreement with those of several authors [28,29]. Thereby, under acid conditions, heavy metals may became more available for plant uptake as they were in ion form [22]. The decrease of soil pH can also cause a faster movement of TME from surface horizons to lower depths [1]. However, when the capacity of soils to retain toxic metals is reduced due to changes in pH, metal ions that have a relatively high mobility can migrate in depth and contaminate groundwater by percolation into the soil [11]. In this context, [20] have shown that this decrease of pH values can be explained by the leaching of calcium carbonates responsible for soil alkalinity. [30] found that soil pH decreased following long-term TWW application due to the elevated microbial activity and the consequently higher production of CO_2 in the upper layer. An additional possible mechanism was the oxidation of organic compounds and nitrification of ammonium, which produces acidity. Other researchers found that soil pH increased with wastewater irrigation [31] and they attributed this pH increase to the chemistry and high content of basic cations such as Na, Ca, and Mg in the TWW applied for a long period.

3.4. Effect of irrigation by TWW on soil salinity

The monitoring of soil salinity control is performed by the EC. The EC values increased as we moved from the surface horizons to the deep horizons for both studied soil profiles (Table 3). We noticed important values at depth especially for P1. EC varies from 0.8 mS/cm to 5 mS/cm for P1 and from 1.4 mS/cm to 2.8 mS/cm for P2. The low EC of surface layers of P1 was due to the leaching of salt

Table 2
TME concentrations (mg/L) of the studied soils

	Depth (cm)	Mn	Cr	Pb	Zn	Ni	Cu	Fe	Cd	Co
P1	0–30	29.30	2.70	1.19	0.00	0.00	0.00	0.01	1.20	0.01
	30–60	24.10	2.00	0.20	0.00	0.00	0.00	0.01	0.60	0.02
	60–120	22.40	2.00	0.21	0.00	0.00	0.00	0.07	0.50	0.03
P2	0–30	23.60	3.10	0.21	0.00	0.00	0.00	0.01	0.90	0.03
	30–60	23.70	2.50	0.20	0.00	0.00	0.00	0.00	0.80	0.04
	60–120	26.10	17.70	0.22	0.33	0.00	0.00	0.02	0.90	0.04

Mn: manganese, Cr: chromium, Pb: lead, Zn: zinc, Ni: nickel, Cu: copper, Fe: iron, Cd: cadmium, Co: cobalt

Table 3
Selected physico-chemical properties of the studied soils

	Depth (cm)	pH	EC (mS/cm)	ESP %	Clay %	Silt %	Sand %	$CaCO_3$T %	TOC %	TN %	C/N
P1	0–30	7.1	0.8	1.54	51	33	13	34	0.92	1.23	7.47
	30–60	7.7	1.5	3.62	53	32	13	38	0.85	0.86	9.79
	60–120	7.5	5	8.40	48	31	19	35	2.13	0.60	33.80
P2	0–30	7.4	1.4	7.63	48	29	20	27	1.01	0.95	10.60
	30–60	7.5	1.5	2.87	53	32	12	27	1.13	0.78	14.41
	60–120	7.2	2.8	5.62	53	33	12	30	0.68	0.72	9.34

EC: Electrical Conductivity, ESP: the exchangeable sodium percentage, $CaCO_3$T: lime, TOC: Total Organic Carbon, TN: Total Nitrogen

by the TWW. Indeed, vertical distribution of salt in soil was directly related to the water movements, which were governed by the rate of irrigation [32]. The increased salinity at the bottom of P1 was due to the irrigation for a long time by TWW that has high EC close to 4 mS/cm and to the accumulation of salts resulted of evaporation and capillary rise. Other workers have also reported similar observations [4,33]. Indeed, they showed a significant increase of soil salinity according to the length of the period of the wastewater application. In 2005, [34] found that in the soil surface horizons of Cebala-Borj Touil irrigated perimeter, salinity was on average between 2 mS/cm and 4 mS/cm but varies between 4 mS/cm and 6 mS/cm in depth. The author explained this phenomenon by the rise of the saline water table that controls soils salinity in this area. These results prove that salinity was about to settle mainly in depth. In addition, it was important to recall that the soils of the studied perimeter were developed in the depression "Garaat Ben Ammar" on alluvial deposits of marine origin, already of high salt content. It was, therefore, a primary salinization, getting worse by a secondary salinization caused by irrigation with TWW. The increase in salinity in this range was also amplified by a low internal drainage of the soil due to heavy clay texture, which renders these soils more vulnerable to salinization [35]. Value of the exchangeable sodium percentage (ESP) was also important in the deep soil layer (8.40%) of P1 compared to P2 (5.62%). Although it has not exceeded 15%, this value may presented a risk for soil sodication. Indeed, sodicity problems arise slowly, but once soil deterioration occur within short times, sodic soils are rarely remedied [32]. Thus, the unbalance between monovalent and divalent cations causes swelling and shrinkage in soils with a certain quantity of clay. When this problem become evident, it can provoke a major non-permeable behavior that does not allow water to enter the soil.

3.5. Effect of irrigation by TWW on soil OM content

The TOC levels were relatively low; they seem to be preferentially localized in the surface horizons of the two studied soils P1 and P2. The TOC percentages vary from 0.92% to 2.13% in the irrigated soil and from 1.01% to 0.68% in the control (Table 3). The vertical distribution of TN has the same behavior as the TOC. TN content drops considerably according to depth. It varies from 1.23% to 0.63% and from 0.95% to 0.72% respectively for P1 and P2 (Table 3). In the case of P1, the TOC decreased in the top layer and recorded an increase in depth. This may explain the slight difference in the soil color of P1 that became clearer on the surface and darker in deep horizons compared to the control. These results are in line with some previous findings [21,25] and can be attributed to the accumulation of organic materials brought by TWW. Indeed, the TSS in TWW can include algal particles, which enrich the soil with OM and nutrients after their biodegradation. According to [30], a long-term TWW irrigation could be of agricultural interest mainly due to its OM content, but micronutrient concentrations in the surface horizons can be negatively affected because complexing of metals favored their transport throughout the soil profile, which may eventually lead to the deterioration of groundwater quality and micronutrient deficiency. In the control (P2), the C/N ratios were high at the surface and decreased with depth. Conversely, there has been an increase in the C/N ratio in the deeper horizons of the irrigated soil (P1). The values increased from 7 to 33 (Table 3). The high value of C/N ratio at the bottom of P1 clearly indicates that the OM accumulated in the deep horizon was humified. This is may be due to the high salinity of the irrigation water (EC≈4 mS/cm) which contributes to inhibit the microbiological activity that was responsible for the degradation and the mineralization of OM [36]. Besides, the presence of hydromorphic conditions in this irrigated area prevents OM degradation and favorites humification.

4. Conclusion

The use of TWW for agricultural irrigation becomes increasingly important in water stressed regions for substituting fresh water resources. This practice has potential adverse effects on soil quality mainly with the continued use of TWW. Our research can contribute to the evaluation of long-term effects of TWW on soil properties based on a study case from Cebela-Borj Touil irrigated perimeter (North Tunisia). Our results showed that TWW used for irrigation presented a high organic load, an alkaline pH, a relatively high salinity (3.73 mS/cm) and TME concentration under the detection limit. After 25 years of irrigation applied by flooding through furrows, we found noticeable differences in some soil properties as a result of TWW application. The comparison of two profiles, an irrigated soil with TWW and a control (non-irrigated soil) showed that the irrigated soil presented a different structure, a lighter color and a slightly finer texture, mainly in surface layers than the control. Besides, a mild acidification of soil surface and a problem of salinization/sodication accompanied with humified OM accumulation in soil deep horizons were observed for the soil irrigated with TWW. The log-term irrigation with TWW did not affect TME concentrations in the irrigated soil except for Mn for which an enrichment of the top horizon was observed. Further, the poor quality of TWW used in our studied area, could stand for an imminent threat for the soil fertility and possibly for the quality of free water table. Indeed, the results of this study clearly showed that TWW reuse in irrigation must be conditioned by some management measures (crop selection, irrigation methods and procedures) and may be by the implantation of improved tertiary treatment technologies, which are not currently used in Tunisia.

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