Desalination and Water Treatment
www.deswater.com

doi: 10.5004/dwt.2018.21825

102 (2018) 61–70
January

Export of salt and heavy metals in an area irrigated with treated wastewater: a case study from Cebala Borj-Touil (Tunisia)

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Received 27 February 2017; Accepted 17 December 2017

ABSTRACT

The Cebala area faces serious problems of salinization caused by shallow saline groundwater and bad drainage system. Situated in northern Tunisia, it is considered the largest area irrigated with treated wastewater in Tunisia. To analyze the environmental impact of the reuse of treated wastewater, an experimental plot was installed in a secondary open drainage for more than 2 years. In this paper, we present the main experimental results: an analysis of volume and the salinity of drained water, a characterization of chemical parameters and heavy metals in drainage water and the exported quantity of salts and heavy metals. From July 2013 to January 2016, we measured in 36 ha, a total sum of 985 m^3 ha^{-1} d^{-1} drained water with an average flow of 1.07 m^3 ha^{-1} d^{-1} and an average total dissolved solids (TDS) of 6 g L^{-1}. The characterization of the drainage water shows a neutral to slightly alkaline pH of water. The chemical compositions of soluble salts show that the drainage water is dominated by the sodium and the chloride. The heavy metals Zn, Ni, Fe and Cr were the most abundant elements in the drainage ditch, whereas Cu and Cd were the less abundant ones. Compared with the Tunisian standard for the discharge of treated wastewater NT 106.02, both Cr and Cd concentrations constantly exceed it. This paper shows that special attention needs to be given to the drainage system in Cebala. This is due to the accumulation of 214 tons of slat in 36 million m^3 drainage water through the drainage ditch D2. The exported mass of heavy metal in the sampling site was in decreasing order of: Cr > Zn > Ni > Co > Pb > Mn > Cd > Fe > Cu. However, further studies are needed to assess their long-term potential risk on the environment.

Keywords: Treated wastewater; Heavy metals; Salinity; Drainage water; Ecosystem

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1. Introduction

Because irrigation is vital for agricultural production, the growing demand of water for agricultural irrigation has caused a significant increase of treated and/or untreated wastewater worldwide [1].

Agricultural wastewater reuse is a major element of water resources development and management that provides alternative and innovatory options for agriculture [2]. The reuse of reclaimed water for irrigation enhances agricultural productivity by providing water and nutrients, and improving

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crop yields [3–5]. It also reduces the use of chemical fertilizers [6,7]. However, despite the benefits, the overuse and the misuse of water in irrigated agriculture could potentially increase the risk of surface water and groundwater contamination [7–9]. According to FAO [10], agricultural water reuse has resulted in large-scale waterlogging and salinity; also accounts for the pollution of freshwater resources through deep percolation that may cause severe environmental pollution and threaten public health.

In North African countries, irrigated agriculture is strongly influenced by poor water quality (saline or polluted) and several other problems related to agricultural practices such as the lack of drainage, as well as shallow saline water tables, and salinization of soil and groundwater [11]. Soil salinization is also caused by inappropriate crop management, such as wrong amount and schedule of irrigation, unsuitable irrigation systems or from insufficient drainage [12]. Without proper drainage, salts accumulate in the upper soil profile, especially in combination with high evapotranspiration and insufficient leaching [1]. On the global scale, nearly 50% of the total irrigated lands in arid and semi-arid regions have soil salinization problems [13]. Soil salinity reduces the mineralization of organic carbon. The release of CO_2 is inversely proportional to the salinity of the water supplied. Generally, dynamic equilibrium conditions for groundwater and soil salinity are reached when the incoming salt load added by irrigation water equals the outgoing salt load removed by the natural and artificial drainage waters. However, the water table rises gradually when there is an imbalance between percolating water and drainage water, until a dynamic equilibrium is reached [14]. Nevertheless, due to the reduction in water percolation and the increase in capillary rise, high water tables usually leads to salinization of shallow groundwater and soil [15].

The mitigation and the control of soil salinity is one of the main challenges in the agriculture of the 21st century, particularly on irrigated soils [16]. In Tunisia, several irrigated areas are affected by land degradation and a decrease of soil productivity. To combat this problem, drainage networks have been installed. The most affected areas are the Medjerda valley in the north, the Kairouan plain in the center and the oases in the south [17]. These old perimeters were not drained for a long time.

The experimental site for this study is located at Cebala Borj-Touil. It is one of the most vulnerable areas to pollution influenced by the use of treated saline wastewater in combination with a saline and shallow water table [18,19]. This perimeter is considered as an example of a complex, large area by salinization and primary waterlogging [20]. Therefore, the prevention of degradation in the Cebala Borj-Touil water quality requires effective monitoring of several heavy metal parameters. Specific papers about the Cebala Borj-Touil perimeter are scarce because of weak interest granted to this area characterized by poor drainage and bad groundwater quality. Furthermore, few investigators have worked on heavy metal pollution in Tunisia. In fact, there is no report about the long-term environmental risk of the use of treated wastewater on drainage water in this area. Hence, in order to evaluate spatial and temporal variations in salts and total heavy metals pollution, this study was needed. The aims of this investigation were: (i) to determine volume and salinity of drainage water of Cebala Borj-Touil, (ii) to analyze the chemical composition of drainage water and (iii) to assess the exported amount of salts and heavy metals (Pb, Cu, Co, Cd, Zn, Fe, Ni, Cr and Mn).

2. Materials and methods

2.1. Monitoring area

The research site is located at Cebala Borj-Touil perimeter, the most important agricultural perimeter in Tunisia with an area of 3,200 ha agricultural land. The perimeter was irrigated with treated wastewater since 1989. It is located in northeastern Tunisia in the suburbs of the capital (36°50 N; 9°75 E). In the east, it is delimited by a depression called Garaat Ben Ammar, in the west by the road GP 8, in the south by the Nahli relief and in the north by the Medjerda wadi. It drains naturally to the Mediterranean Sea. Across the plain there are two ephemeral streams, the Khelij and the El Maleh. The climate is semi-arid with a wet season from September to May and a dry season from June to August, average annual rainfall is 450 mm [21] and potential evaporation measured by the Piche evaporimeter is 1,300 mm. The region is characterized by a shallow saline water table ranging from 0.5 to 7.6 m. The soil is clayey with a degraded structure, it is used mainly for the production of cereal and industrial fodders and drives the internal drainage quite slow [22,23]. Indeed, some farming restrictions were imposed by the government to avoid sanitary and health risks. That is why only fodders, cereals and industrial products are imposed as crop farming, because they are low profitability products [22]. The situation is getting worse because the drainage network installed two decades ago is degrading [20].

The drainage system of the Cebala Borj-Touil perimeter consists of an open ditch network covering almost the entire area and tile drainage on 500 ha (Fig. 1). Six main collectors drain the perimeter: The first one is 1.2 m deep carrier canal drains about 300 ha and collects the water of six major collectors; the El Khlij canal drains 430 ha, but it has a very low slope (less than 0.02%) and is almost completely clogged by reed and weed; the main axis of the drainage network is the canal principal II (CPII) which drains the most important part of the perimeter of about 1,450 ha. This area suffers from high groundwater levels, particularly downstream of the CPII. The fourth is the Prisonniers canal; it drains an area of 703 ha and receives the drainage water of wadi El Khalij and the CPII. The fifth and the sixth are the El Maleh and Khelij wadis draining 560 and 900 ha, respectively.

Table 1 shows the average composition of the treated wastewater. Sixteen water samples were collected periodically during the experimental period.

2.2. Experimental setup and field measurements

Since no information was available about the water of the drainage ditch, a station was installed at the secondary open drainage D2 to analyze the environmental impact of the irrigation with treated wastewater, it drains 36 ha.

- To measure discharge and salinity of the drainage water, a Pollubac device was installed at the outlet of the drainage ditch (D2). Pollubac is a discharge trough operating

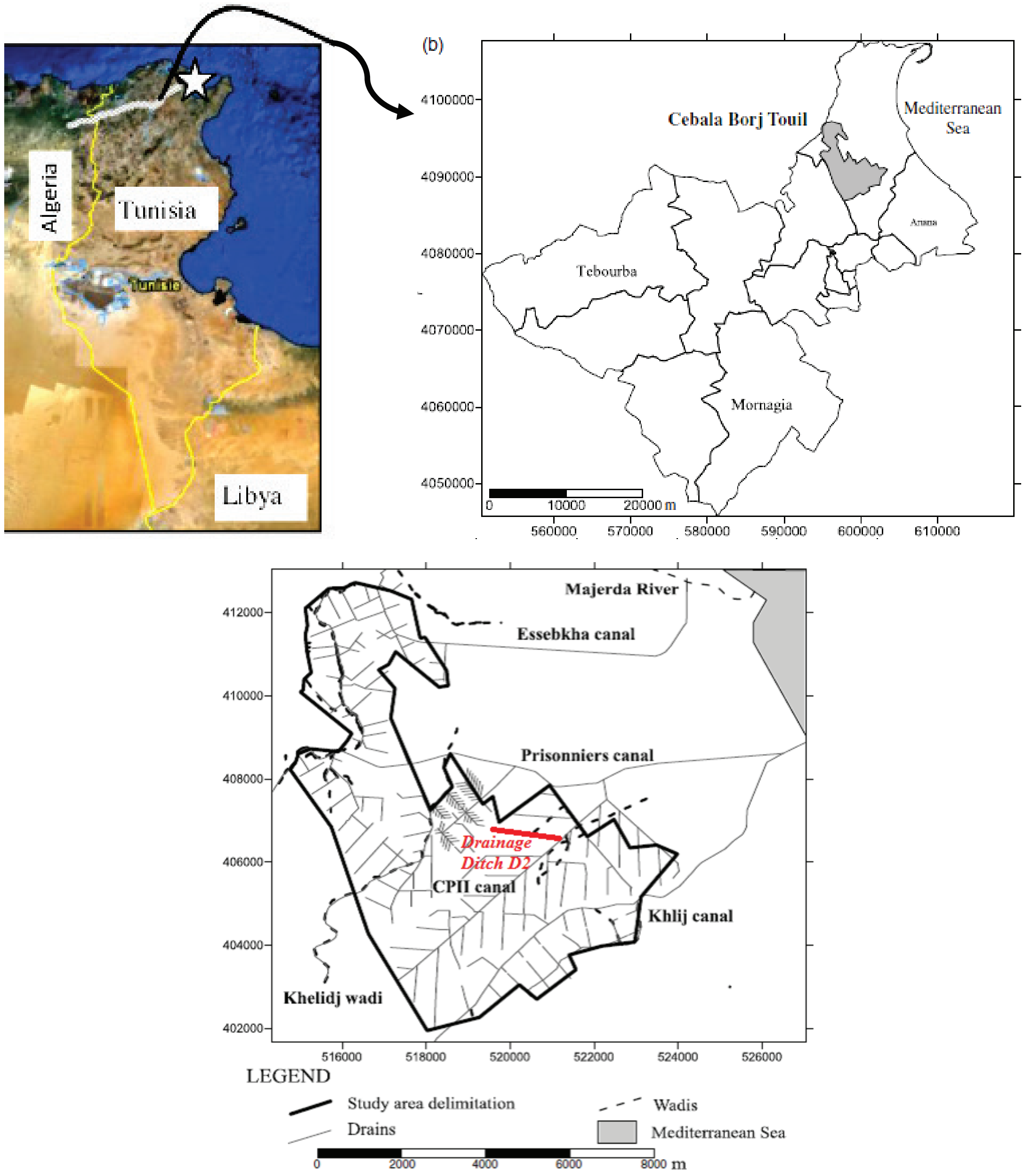

Fig. 1. Location of the research site within Cebala Borj-Touil, Tunisia.

similar to the discharge weirs used in hydraulic laboratories. The 1,100 × 600 × 350 (LWH) measures discharge (Q) up to 8 L s^{-1}. It consists of an upstream basin with an energy absorbing deflector, a downstream basin with a removable stilling well, a weir with compound filled edges, a lateral measurement chamber with a scale and two lateral water level measurement points [24]. The water level is measured at the triangular weir. For each weir there is a standard calibration curve (Fig. 2).

- To estimate the amount of exported salts, a calibrated sensor for water level and electrical conductivity (EC; CTD-Diver 5610) was installed from July 2013 to January 2016.
- The CTD-Diver is a compact sensor for water level, temperature and conductivity (measuring range 0–120 mS cm^{-1}). It has a ceramic housing with a diameter of 2.2 cm and length of 13.5 cm. The CTD-Diver can record the piezometric level and the salinity with a time interval of 4 h. With an additional sensor for barometric pressure, the influence of atmospheric pressure can be removed with the use Diver-Office software (Diver-Office 2008) [25].

The CTD-Diver recordings were validated with manual measurements of water level (H) and EC. The validation results are satisfactory with a high correlation coefficient varying between 0.7 and 0.9 [26]. The correlation coefficient R^2 was 0.94 for water level and R^2 = 0.9 for conductivity.

The EC (in dS m^{-1} at 25°C) of the drainage water was converted to the TDS (g L^{-1}) by using a constant factor of 0.64 [27].

The total amount of salt exported may be calculated from the volume of water pumped from the drainage ditch: M_{salt} (kg) = ($TDS_j \times Q$)/1,000.

Table 1
Descriptive statistics of TWW chemistry data in Cebala (n = 16)

	Min.	Max.	Average	SD	CV
pH	6.670	8.260	7.700	0.58	7.60
EC, dS m^{-1}	2.860	7.600	4.180	1.55	37.15
Na^+, meq L^{-1}	12.560	48.340	21.780	10.49	48.16
K^+, meq L^{-1}	0.380	1.140	0.920	0.30	32.53
Ca^{2+}, meq L^{-1}	2.000	6.000	3.170	1.25	39.47
Mg^{2+}, meq L^{-1}	3.000	8.000	5.330	1.52	28.51
HCO_3^-, meq L^{-1}	6.500	20.500	10.110	4.13	40.84
Cl^-, meq L^{-1}	16.920	31.020	20.840	4.28	20.52
SO_4^{2-}, meq L^{-1}	4.510	13.350	8.860	3.09	34.90
Cd, mg L^{-1}	0.001	0.010	0.010	0.00	50.55
Co, mg L^{-1}	0.015	0.028	0.020	0.00	21.20
Cr, mg L^{-1}	0.000	0.060	0.030	0.03	102.50
Cu, mg L^{-1}	0.010	0.061	0.020	0.02	80.64
Fe, mg L^{-1}	0.000	0.855	0.250	0.32	129.68
Mn, mg L^{-1}	0.035	0.412	0.110	0.15	135.88
Ni, mg L^{-1}	0.002	0.048	0.030	0.02	54.60
Pb, mg L^{-1}	0.000	0.048	0.030	0.02	72.26
Zn, mg L^{-1}	0.006	0.187	0.120	0.07	56.99

Notes: Min., minimum; Max., maximum; SD, standard deviation; CV, coefficient of variation (%).

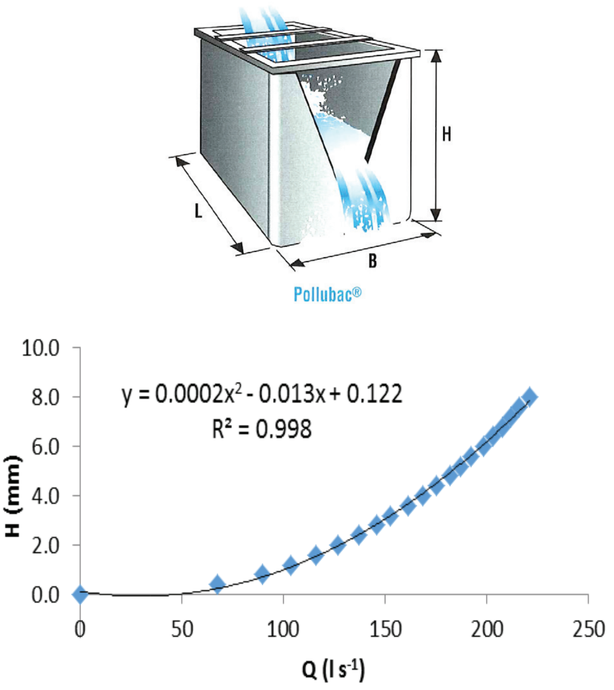

Fig. 2. Calibration curve of the Pollubac.

The volumes of drained water and the concentrations of heavy metals in the drainage water were used to calculate the annual heavy metal exporting rates.

2.3. Analytical methods

Samples of drainage water were collected from the drainage outlet D2. The following parameters were analyzed: pH, temperature and EC with a conductivity meter type Cond level 2 WTW inolab. The different elements were analyzed with the following methods: sodium (Na^+) and potassium (K^+) with flame emission spectroscopy (NF-A20-603); calcium (Ca^{2+}) and magnesium with versenate titration method (EDTA). Bicarbonate (HCO_3^-) concentration with titration with sulfuric acid (H_2SO_4) and chloride (Cl^-) concentration with titration with silver nitrate ($AgNO_3$). Trace elements such as Cr, Co, Fe, Cu, Mn, Zn, Pb, Cd and Ni were measured using atomic absorption spectrometry according to standard methods AFNOR N°NF FD T90-112 (1998).

2.4. Statistical analysis

Statistical analyses included calculation of basic descriptive statistics average, minimum, maximum, standard deviation and coefficient of variation (CV). The interpretation of the CV was based on the following limits: CV < 10%: low variability;10% < CV < 50%: average variability and CV > 100%: high variability.

3. Results and discussion

3.1. Discharge

3.1.1. Variation of flow

Fig. 3 shows daily discharge and precipitation values from July 2013 until January 2016. The average discharge is 1.07 m^3 ha^{-1} d^{-1} and the total amount for the whole period 985 m^3 ha^{-1} during the whole experiment.

Toward mid-October, the water flow increases up to 2.774 m^3 ha^{-1} d^{-1}. A similar peak with 1.644 m^3 ha^{-1} d^{-1} was recorded at 14/03/2015. It can be explained by the high rainfall and low evapotranspiration during that period. The high values of 2.025 m^3 ha^{-1} d^{-1} in April 2014 and 2.08 m^3 ha^{-1} d^{-1} in May 2015 are related to irrigation effect. This is partly explained by the lower irrigation rate in the Cebala area.

These variations are controlled by several factors. The depth of the groundwater is strongly influenced by climatic conditions such as scarcity or abundance of rainfall. The drainage conditions are also an important factor because they correspond to the initial development of the drainage network and its degradation, in addition to the periodic maintenance, which usually occurs once in every 3 years [20].

3.1.2. Drainage water salinity

Fig. 4 displays the monthly salinity during the experiment. The change in salinity had different trends in both years. The average was 6.0 g L^{-1} with a range from 3.8 to 7.8 g L^{-1} (CV = 15%).

Maximum concentrations were reached in summer and minimum concentrations in winter because of the dilution

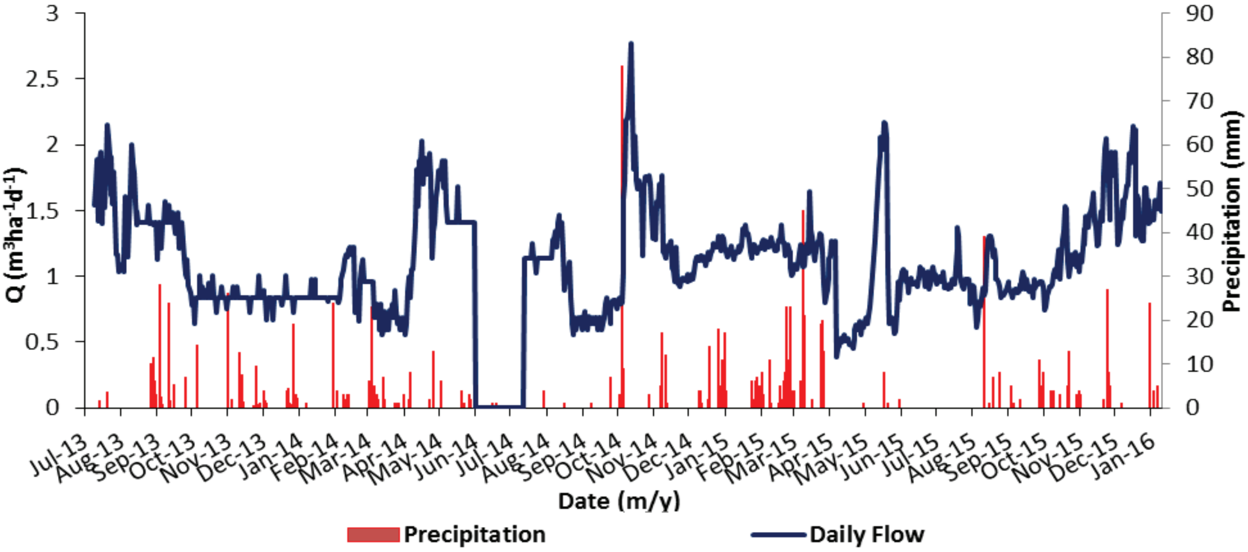

Fig. 3. Daily flow variation at the drainage ditch in irrigated area of Cebala Borj-Touil from 09/07/2013 to 09/01/2016.

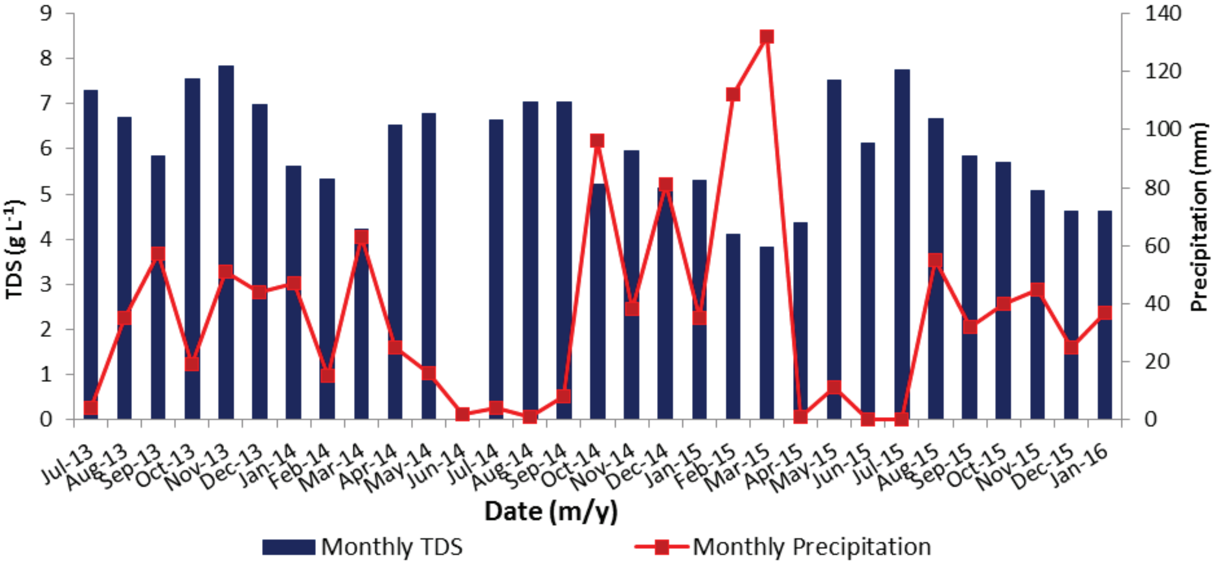

Fig. 4. Temporal variation of salinity at the drainage ditch from July 2013 to January 2016.

of salts by precipitation. Thus, winter leaching effect is more efficient than irrigation [28]. As illustrated in Fig. 5, there is a strong positive correlation between drainage salinity and precipitation with a correlation coefficient of 0.8087. In fact during rainfall events, salt concentration in the drainage water decreased systematically and increased during recession [29]. Magesan et al. [30] and Arlot [31] explained that the salt content in the drainage system can be controlled by two typical reactions depending on the origin of the solute: (i) When the saline irrigation water is applied at the soil surface, the concentration in the drainage water increases with increasing flow. (ii) When the solute originates from the groundwater, the concentration in the drainage water decreases with increasing flow.

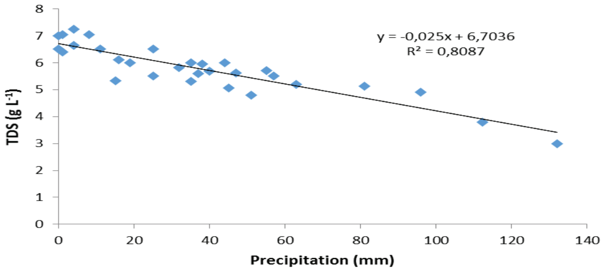

Fig. 5. Correlation figure of drainage salinity against precipitation.

3.2. *Quality of drainage water*

3.2.1. *Chemical composition*

Table 2 shows the average dissolved concentration of each chemical component in the drainage water. pH ranges from a neutral value of 7.26 to slightly alkaline with 8.54, the exceptionally high pH values were likely related to alkaline irrigation water (Table 1). EC can be used to evaluate the total soluble salt concentration of inorganic substances. Average EC was increased with a maximum of 13.5 dS m^{-1} and a minimum of 4 dS m^{-1}. The chemical analysis of the soluble salts shows that the drainage water is dominated by sodium and chloride.

3.2.2. *Heavy metal composition*

Range (min–max), mean and coefficient variation of the heavy metals Pb, Cu, Zn, Fe, Ni, Cr, Cd, Co and Mn concentrations in 30 drainage water were analyzed (Table 3).

Zn, Ni, Fe and Cr were the most abundant elements in the drainage ditch, whereas Cu and Cd were less abundant ones. All elements exhibit a high seasonal variation as reflected by the high CV [32]. Among the nine heavy metals analyzed, Zn was most abundant in the drainage ditch with a mean level of 0.08 mg L^{-1}. The mean concentration of Zn ranged from 0.011 to 0.78 mg L^{-1}. The high levels of Zn suggest that wastewater is the source of the pollution since the amount of Zn in irrigation water is 0.120 mg L^{-1} (Table 1). As mentioned in De Zuane [33], Zn is the most abundant element found in the earth's crust. Zn can enter the aquatic environment from a number of sources such as industrial discharges, sewage effluents and terrestrial runoff [34].

Concentration of Fe varied between 0.041 and 0.317 mg L^{-1}. The distribution of the iron was approximately similar to Cr. The mean concentration of Ni ranged from 0.012 to 0.155 mg L^{-1} However, Ni is easily accumulated in phytoplankton or other aquatic plants and it can be deposited in the sediment by many processes such as precipitation, complexation and adsorption on clay particles [35]. The concentration of Pb ranged from 0.01 to 0.093 mg L^{-1}. Cu content ranged from 0.001 to 0.066 mg L^{-1}. Regarding the irrigation water, the concentrations of Pb and Cu were not high (Table 1). This clearly indicated that anthropogenic input could be the source of Pb and Cu. The concentrations of Cr, Co, Cd ranged between 0 and 0.196 mg L^{-1}, 0.005 and 0.179 mg L^{-1}, 0.005 and 0.027 mg L^{-1}, respectively.

The temporal variations of heavy metals are analyzed and compared with the Tunisian standard NT 106.02 (Fig. 6).

The concentrations of Pb, Mn, Zn, Fe, Ni, and Cu were the lowest. Both Cr and Cd concentrations in the ditch water exceeded the Tunisian standard. Cd in drainage water varies widely between 0.025 mg L^{-1} and 0.005 mg L^{-1}. In fact, 24 of the 26 Cd samples exceed the Tunisian Standard. The amount of Cr constantly exceeds the Tunisian threshold values of 0.01 mg L^{-1}. Co contents are less variable with values between 0.17 and 0.005 mg L^{-1}. They exceed the Tunisian standard (0.1 mg L^{-1}) between November and December 2014.

Pb varies between 0.23 and about 0.01 mg L^{-1}. It highly exceeds Tunisian standard in winter (0.1 mg L^{-1}). Levels of Mn, Zn, Fe and Cu are much lower than the Tunisian threshold. The amount of Ni varies between 0.15 and 0.01 mg L^{-1}.

Table 2
Statistics of drainage water quality element analysis in Cebala ($n = 42$)

	Min.	Max.	Average	CV
pH	7.260	8.540	7.860	7.640
EC, dS m^{-1}	4.000	13.500	9.700	34.420
Na^+, g L^{-1}	0.342	3.326	1.894	32.460
K^+, g L^{-1}	0.001	0.352	0.042	167.740
Ca^{2+}, g L^{-1}	0.044	0.240	0.126	39.220
Mg^{2+}, g L^{-1}	0.001	0.201	0.042	111.930
HCO_3^-, g L^{-1}	0.490	1.255	0.676	22.730
Cl^-, g L^{-1}	0.550	4.400	2.700	24.280
SO_4^{2-}, g L^{-1}	0.105	1.387	0.727	41.520

Notes: Min., minimum; Max., maximum; CV, coefficient of variation (%)

Table 3
Statistics of metal concentrations

	Min.	Max.	Mean	CV	NT 106.02
Cd	0.005	0.027	0.014	38.070	0.005
Co	0.005	0.179	0.052	62.950	0.100
Cr	0.000	0.196	0.079	56.340	0.010
Cu	0.001	0.066	0.012	113.520	0.500
Fe	0.041	0.317	0.147	51.270	1.000
Mn	0.000	0.153	0.043	88.410	0.500
Ni	0.012	0.155	0.070	45.640	0.200
Pb	0.010	0.093	0.046	62.520	0.100
Zn	0.011	0.780	0.080	190.050	5.000

Notes: Min., minimum; Max., maximum; CV, coefficient of variation (%). All concentrations in mg L^{-1}.

The seasonal variations of all heavy metals are quite high (Fig. 6) with the maximum of Pb, Fe, Zn, Cu and Mn in autumn (September); Ni, Co and Cr in winter and Cd in summer. The seasonal variations of Cu, Zn, Mn and Fe were small. The low concentrations of the elements in the ditch water could be explained by their low accumulation in the soils on adjacent fields and the high soil pH values (pH > 7) which decreases the solubility and therefore their release from the soils [36].

According to Li et al. [37], the discharge of wastewater can pollute the entire study area. The high level of Cd in summer could be attributed to the intense evaporation. In the wet seasons (autumn and winter), dilution by high precipitation was the most plausible explanation for the low heavy metals concentrations [38]. This result is consistent with other studies [39,40]. The high Cr concentrations in the ditch water can be considered as the greatest threats to organisms in the sediments [37].

3.3. *Exported salt and heavy metals*

Between July 2013 and January 2016, the discharge of 36 million m^3 exported 214.17 tons of salts from a total area of 36 ha with an average of 6 kg ha^{-1} d^{-1} (Fig. 7).

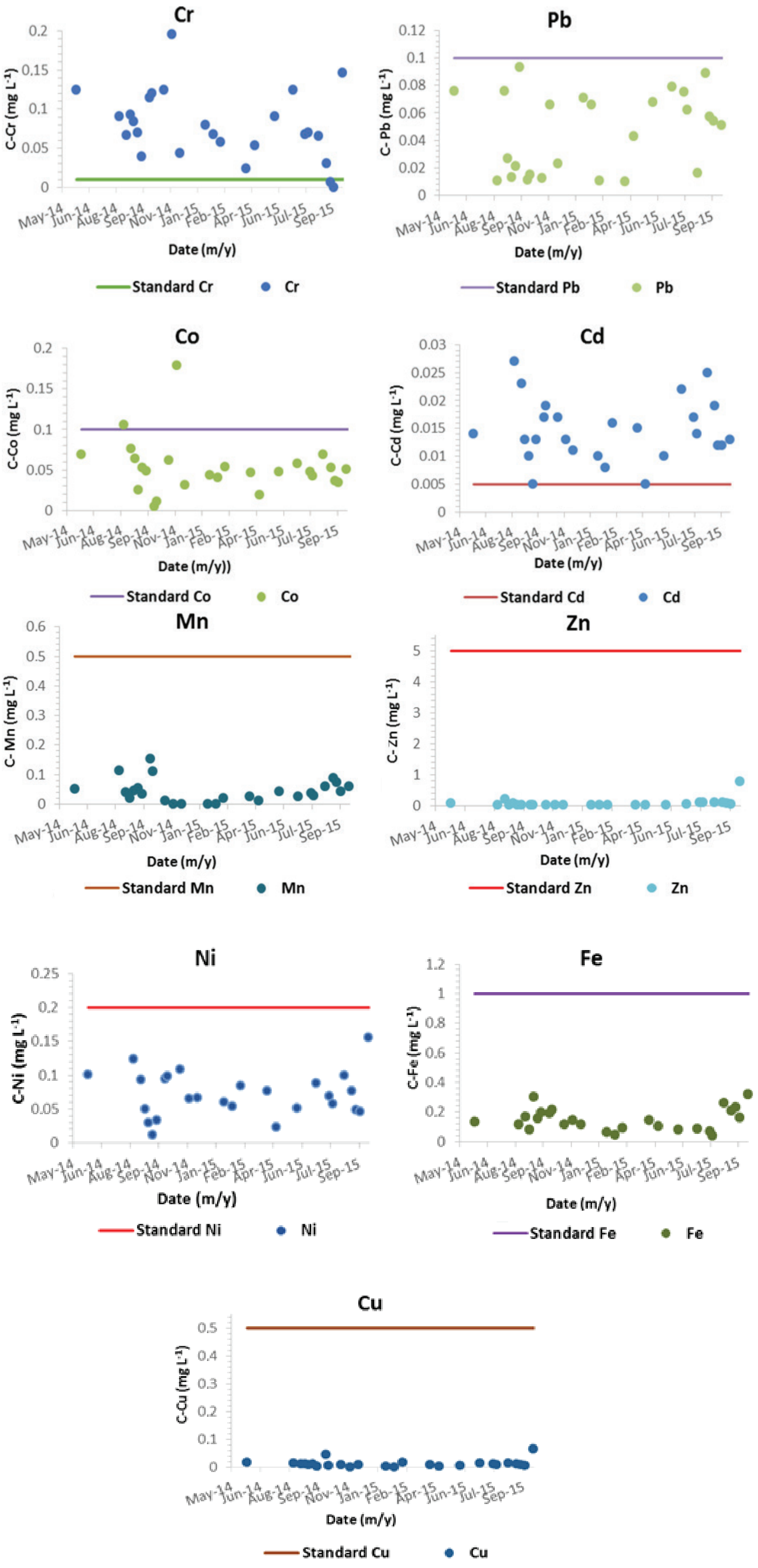

Fig. 6. Temporal variations of heavy metals concentrations in the drainage ditch.

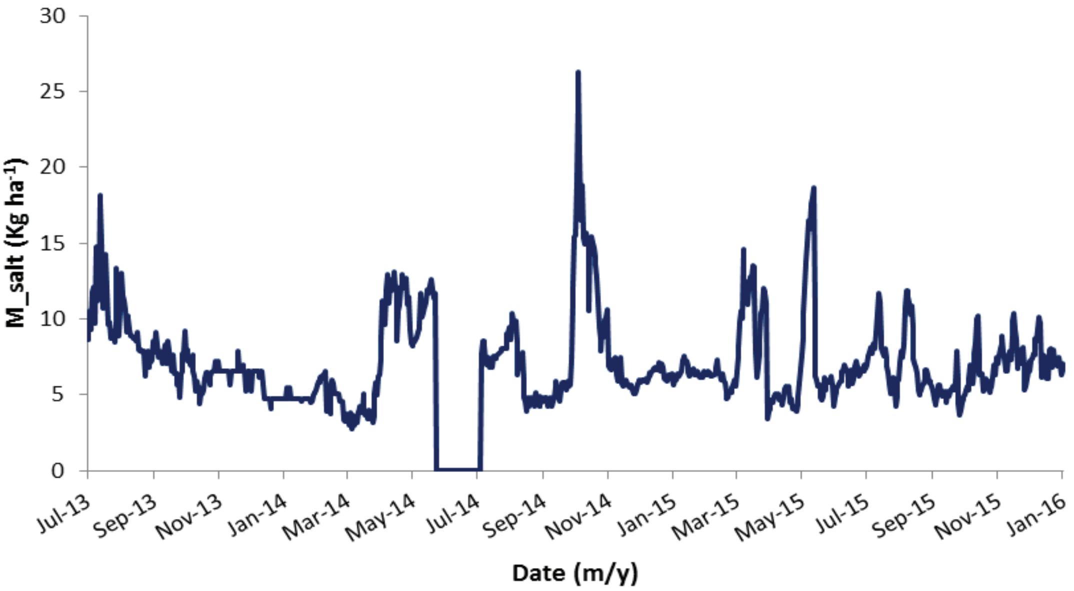

Fig. 7. Amount of salts exported with the drainage water.

Table 4 summarizes the exported amount of nine metals. During two and a half years, the drainage water deplenish significant amounts of heavy metals. The amounts were lowest for Cd, Fe and Cu and highest for Cr, Zn and Ni. The relative abundance of heavy metals in the sampling site was in decreasing order of: Cr > Zn > Ni > Co > Pb > Mn > Cd > Fe > Cu.

As depicted in Table 4, the drainage system exported 71.11 kg of Cr, 67.66 kg of Zn and 65.59 kg of Ni. These high amounts are probably related to the accumulation in the soil of adjacent fields [41]. The leached water from soils irrigated with treated wastewater causes contamination of groundwater, especially during rainy periods. It is then transmitted to drainage ditches, reaches the sea and affects the marine ecosystem. Robertson and Taylor [42] also reported that the contaminants are mobilized and transported to drainage systems after storms, thus affecting water quality [42].

Table 4
Salts removed by drainage between July 2013 and January 2016

Elements	Salt removed	Min.	Max.	Average	CV
Cd	13.00	0.106	1.152	0.520	52
Co	47.23	0.159	6.072	1.810	75
Cr	71.11	0.000	0.785	2.730	68
Cu	10.76	0.033	2.197	0.414	116
Fe	12.47	1.289	10.719	4.797	55
Mn	38.21	0.000	4.893	1.469	98
Ni	65.59	0.278	6.308	2.522	61
Pb	40.87	0.143	4.743	1.571	74
Zn	64.44	0.404	25.967	2.593	192

Notes: Min., minimum; Max., maximum; CV, coefficient of variation (%). All concentrations in kg.

4. Conclusion

The reuse of wastewater for irrigation became a common practice in semi-arid and arid countries such as Tunisia. The Cebala Borj-Touil perimeter in Northern Tunisia is one of the largest and the most vulnerable areas to pollution; it is already affected by the use of treated saline wastewater. In order to investigate the environmental impact of the use of this practice, we analyzed the water quality in the drainage ditch for an extended period.

During the experiment, 214.17 tons of salt were exported. The heavy metal concentrations vary in time and space. Metals such as Co, Cr and Cd exceed the Tunisian threshold values NT 106.02 and therefore must be controlled. The leaching of the metallic elements takes place especially during rainy periods.

We recommend that urgent actions must be taken at both administrative and technical levels to prevent the aggravation of the problem. These technical actions should been carried out without forgetting to change agricultural practices (crops more tolerant to salinity). We need to improve irrigation efficiency using surface irrigation to reduce salinity problems and water losses on farmers' plots. We also need to limit the deterioration of groundwater characteristics by the deepening of the principal canal (CPII) of the drainage system, and amplifying the maintenance operations of the actual drainage system [20]. Hence, to overcome the above-described problems, the training of personnel involved in Research and Development and of private organism specialists in drainage is a priority.

Furthermore, the study suggests that the waste management in the Cebala Borj-Touil perimeter should be more effective in order to sustain the ecological integrity. On this basis, the agricultural use of treated wastewater requires, even if thresholds levels are respected, a continuous, and long-term monitoring of the entire system from the parcel to the sea.

Acknowledgments

This study was carried out in the Research Laboratory of Non-Conventional Waters Use. It is supported by the Tunisian Ministry of High Education and Scientific Research and the German Ministry of Research (BMBF) in the project INRGREF/TUNGER, and the INRGREF/ACSAD convention.

Symbols

TWW — Treated wastewater
TDS — Total dissolved solids
Q — Water flow
H — Height of water flowing over weir
M_{salt} — Mass of salt

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