IRRIGATION WATER QUALITIES-SOIL POLLUTION (HEAVY METALS AND SALINITY) IN MORNAG IRRIGATED PERIMETER (SW TUNIS, NORTH TUNISIA)

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1. Introduction

On account of continued agricultural development, population growth and economic development, the water demand is constantly increasing but in arid and semi-arid countries natural water resources are limited. Therefore, groundwater resources are becoming increasingly scarce and their quality continues to deteriorate, among other water salinization. So, to remedy this water lack, especially for irrigation which is the biggest water user in these countries, there is recourse (in our case) to different water sources which can have variables physicochemical parameters (pH, salinity and organic matter content). The farmers irrigate by:

- treated wastewater (TWW) which becomes an important agronomic proceeding. This TWW contribute to the recycling of organic matter and restore the soils fertility by the added N.P.K.,
- surface water from dams.

But the nutriments and dissolved compound, in different waters, can be associated to some environment and agronomic problems due to salinization and increasing levels of toxic substances as heavy metals pollutants in soil.

In urban areas, the industrial parks surrounding agriculture zones can be an important pollution sources (Douay et al., 2000). Indeed, the industrial activities produce large quantities of solid and liquid waste which are discharged in situ. But an important waste disposal (among others organic and heavy metal) can be redistributed by wind in agriculture surrounding areas.

To improve agricultural production, many organic and inorganic compounds have been used as a soil amendment to restore the soil fertility (Moore et al., 1995).

The widespread, repeat application and/or wind intake can cause toxic elements increase in soils which will have an effect on the ecological system which can have negative health effect, too (Caldor et al. 2003; Rowbothan et al., 2000).

Soil heavy metals content is not the only parameter for an environmental risk estimation. The mobility, which usually depends on their chemical form, is an important factor for evaluating their distribution and toxicity degree since complex forms are much less available than the free ions (Forstner, 1979). The chemical speciation are closely related to parameters as pH which plays important role in the partition of an element between the dissolved and particle phase. So, the water pH used for irrigation and the pH on sol-residual system are often the most important chemical properties governing trace element sorption, precipitation, solubility and consequently the bioaccumulation and availability of heavy metals pollutants (Kabala and Singh, 2001). The composition and salinity of different waters used for irrigation can play non negligible role in the adsorption or fixing of the pollutants (Forstner, 1979;). Different parameters of soil matrix as chemical composition, mineralogy, texture and organic matter yield control, too, the heavy metals bioactivity (Tam and Wong, 1999).

Mornag irrigated perimeters covers the plain of this region. It is in the northern Tunisian part and 20 km far from Tunis City. This area is known by important agricultural and industrial activities.

Its aquifer has been the main water resource of this region. To limit the overexploitation, there is a creation, in Naasen (Naa) and Irrisaala (IRR) regions irrigated perimeters by treated wastewater (TWW). However the irrigation periods by unconventional water are variables. Moreover, in Mornag to improve the hydrodynamics and water quality of local groundwater, artificial groundwater refill

(AR) by surface water is practiced in the Khlédia area. Everywhere, they irrigated by groundwater and Mejrda-Cap Bon Channel (M-CB.C).

Mornag region includes, too, important industrial parks Ben Arous and Rades, where the solid wastes are an important pollution source (especially heavy metals). These pollutants are indirectly brought to the local agricultural area, mainly by wind.

The water characteristics used for irrigation exchanges from one to other Irrigated perimeter and even for the same perimeter the farmers used different water qualities, we think important to determine the effect of the change in water quality of irrigation on organic matter (TOC), pH and salinity (E.C.) of soil. Our second aim is the evaluation the pollution levels of heavy metals (Ni, Cr, Pb, Cd, Cu, Zn) in surface sediments of Mornag irrigated areas and the contribution of industrial areas and TWW to probably pollution of soil.

2. Geographical framework

The Mornag plain is in the northern plain from Tunisian and 20 km far from Tunis City (Fig. 1). It extends 14 km from the Gulf Tunis (Mediterranean Sea) in the North to Khledia hills in the South. It is limited on the West part by the Rades hills and on the East by the Jebel Er-Rourouf surrounding area (Fig. 1). The climate of the study area is semi-arid to arid with moderate temperature (18°C). The main wind direction in this area is NW and the others are W, SW, SE and NE (CEDA, 2000). The annual rainfall is approximately 450 mm/year (DGRE, 2000). This plain is drained by Meliane river which takes it origin in jbel Bargou and El'Hma. The Morgan aquifer surface is about 200 km² (DGRE, 2000; Lajmi, 1968; Ennabli, 1980). This aquifer system includes (a) the groundwater in recent Quaternary series is composed of clay and clay-detrital complex with silt and sand, especially upstream (Schoeller, 1948), (b) deep aquifer systems are composed of four ancient systems logged in ancient Quaternary, Oligocene, Miocene and Eocene sediments (Lajmi, 1968; Ennabli, 1980).

The unconfined groundwater has been the main water resource of this region. To limit the overexploitation of these resources, (a) since December 1995, the authorities practice the artificial refill (AR) of local groundwater with surface freshwater (M-

CB.C.) by the Khlidia stone-pit which is mainly realized thanks to the good vertical permeability of the recent Quaternary sediments, (b) since 1989, there is creation in a Naasen and Irrisala regions 4 irrigated perimeters by treated wastewater (TWW). In Naasen, we have Sadira and Ouzra which have respectively 230 and 290 hectars. In Irrisala region, there are two irrigated perimeters of which

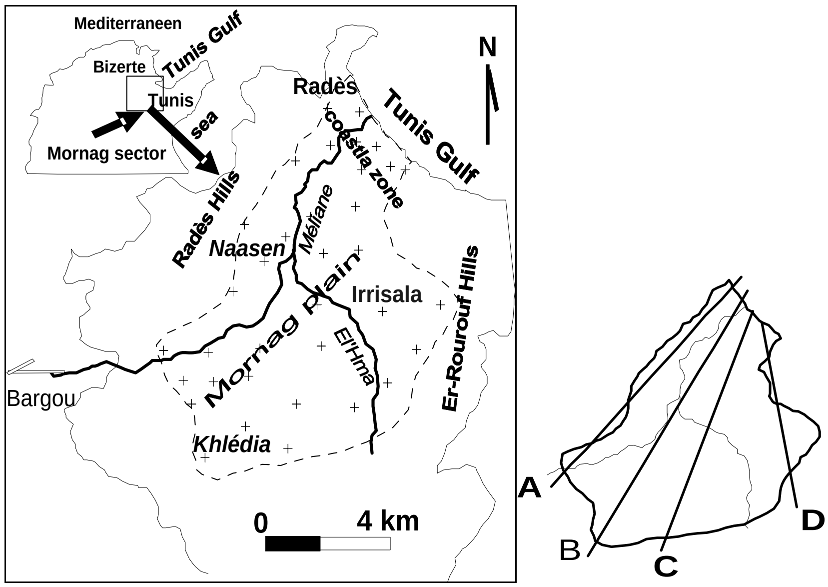

Fig. 1: geographical localization of Mornag plain and the 4 radials
(A, B, C, D) in studied area
- - groundwater limit, + : wells used for pumping groundwater

only one is considered interesting and has a clear history. It covers 180 hectares. In Naassen and Irrisala, farmers have initially irrigated by unconventional water. But five and two years ago respectively in Naassen and Irrisaala, the farmers take back the irrigation, in majority cases, by surface water. Everywhere, they irrigate, in the same time, by surface and ground water.

So, the hydro-chemical water qualities exchange from one sector to another and in the same perimeter.

3. Material and analytical Methods

For water, the pH, salinity and electric conductivity (E.C.) have been measured in situ with pH-meter LPH 230T and conductmeter ORION 150 with platinum electrode.

For sediment, 42 surface soil samples representing the Mornag plain were collected in 2007. Different samples were collected taking into account the macroscopic soil texture, slope and vegetation. For irrigated Naassen and Irrisaala perimeters, all collected samples were initially irrigated by treated wastewater (TWW). In Naassen, samples were also taken from fields irrigated with surface water since 5 years, while samples at Irrisaala were irrigated by Mejrda-Cap Bon channel water only two years ago.

Sediment samples were air-dried to constant weigh (Davison et al., 1998). In a soil-water suspension (10 mg of soil in 50 mL of distilled water) and after two hours of stirring (Montoroi, 1997), the pH and the electric conductivity were measured. The total organic carbon (TOC) has been analyzed by ANNE method. For Ni, Cr, Pb, Cd, Cu, Zn analysis, 0.5 g of soil have been attacked by a mixture of 5 mL of HF (40%), 1.5 mL of $HClO_4$ (70%), 3.75 mL of HCl (37%) and 1.25 mL of HNO_3 (65%) in sand bath at 250°C (AFNOR, 1979). Final solutions were diluted with 100 mL of ultra pure water prior to analyses. 20% of samples and the samples which show the extreme concentrations were run in duplicate. The result is accepted only when the deviation is equal or under the detection limit. The reference samples are those of CRPG-standards (Nancy, France). Analyses are done with Atomic Absorption Spectro-Photometry (AAS) VARIO 6 with graphite furnace where the detection limit for Ni, Cr, Pb, Cd, Cu and Zn are respectively 0.19 pg (picogram), 3.6 pg, 2.1 pg, 5 pg 0.75 pg and 3.6 pg

4. Results and discussion

4.1. Water quality (pH, salinity)

In Mornag, they irrigate by several water qualities. Type (a) groundwater usually has a salinity ranging between 1.5 and 3.5 gL-1, but in many cases, salinity is > 5 g.L-1, without traces of organic compound, and pH is near-neutral. Type (b) treated wastewater (TWW) at Naasen and Irrisala has a salinity of about 2.7 g.L-1, pH of 8.3 and is rich in organic compounds. Type (c) Mejerda-Cap Bon (M-CB C) water channel shows salinity from 0.8 gL-1 to 2 gL-1 (depending on water origin) and low organic compounds. Since the channel is with open sky, the pH of irrigation water varies from winter (~7) to summer (<6).

4.1. Granulometric data

In Mornag plain, there are significant soil texture variations. In Naassen and Irrisaala areas, the textures are comparables and enough homogeneous. It are clay-sandy soil with slightly more than half are composed of sand (Table 1).

Table 1: soil texture variations in Mornag

	Sand	**Clay**	**Silt**
Irrisaala			
Min	47.9	12.0	12.0
Max	62.7	26.4	24.9
Mean	57.4	24.9	16.4
Khlidia			
Min	85.5	3.8	1.8
Max	92.2	8.2	27.0
Mean	89.5	5.5	11.2
Rest of Mornag			
Min	33.6	13.9	5.2
Max	63.6	33.8	45.2

In the rest of the plain, the statistical calculation shows that the soil textures are in average comparable to Naassen and Irrisaala soil. In details, the textures are highly variables from one point to another (Table 1).

4.2. pH, Electric conductivity and TOC content of soil

Soil pH values vary between 6.8 and 8.4 with an average of 7.6.

Spatial pH soil distribution shows that the Irrisaala soil has a highest pH with a minimum of 7.9, a maximum of 8.4 and an average of 8.2 and E.C. whose average is 1.15 mS/cm. The Naassen soil pH is slightly lower (mean pH = 7.88) and also its E.C. (0.62 mS/cm). Khlédia zone has neutral pH soil (pH = 6.96) with the lowest E.C. in Mornag plain (0.16 mS/cm). In the other studied field, the average pH is 7.4 while the E.C. is about 0.31 mS/cm. The soil TOC is between 0.10% and 1.40% with an average concentration of 0.63%. The highest TOC concentrations occur in the soil irrigated by TWW.

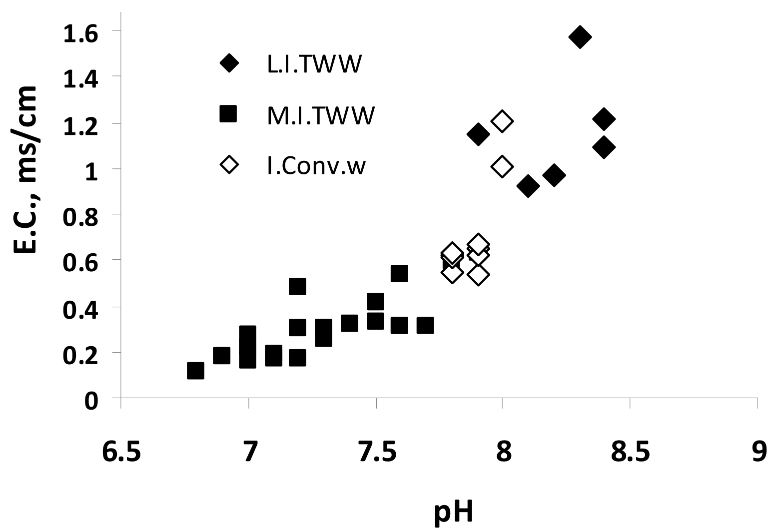

Fig. 2 pH versus E.C. of irrigated perimeters (Irrisaala) for a long time by TWW (L.I.TWW), less time irrigated (Sadira, Ouzra) by TWW (M.I.TWW) and the rest of the perimeters which are irrigated by conventional waters (I.Conv. W)

Figure 2 shows that in Irrisaala the longer irrigation by TWW has the highest pH and E.C.. The take back of irrigation by surface water with lower pH and salinity induced a pH and E.C. soil decrease (M.I.TWW). TWW salinity is relatively high (TWW: ≥ 2.7 gL-1). The used groundwater has also generally higher salinity. Since only the organic content varies from one water type to another, it is likely that organic compounds added to soil by TWW have contributed to increase of pH and E.C. of soil. Indeed, the organic compound of TWW may cause initially the soil pH decrease which can be the result of organic matter decomposition. After, the mineralization of OM in soil reduce the COT tenor (in Mornag from 0.7% to 1.9%) and probably produce carbon and negative charges as hydroxide ions. At the same time, basic cations as K^{+}, Ca^{2+}, Mg^{2+} have liberated (Mohammed and Athamneh, 2004; Mkhabela and Warman, 2005). After, the proton soil adsorption can increase the soil pH (Jardao et al. 2006).

Thereafter, the irrigation by M-CB water channel (lower pH) causes a soil pH decrease (probably proportional to TOC decrease) which explains the virtual positive limited correlation pH-TOC.

Finally, the soil pH and E.C. depend, directly, the pH and the composition of irrigation water and the period of use of treated and fresh water.

4.3. Heavy metals distribution

The average concentrations of heavy metals in surface sediments vary from 1.18 to 113.6 $mg.kg^{-1}$ (Table 2). The abundance order is Ni> Cr> Zn> Pb> Cu> Cd

Table 2. The average and limit values of soil heavy metals content, in mg kg-1

	Ni	**Cr**	**Pb**	**Cd**	**Cu**	**Zn**
I.Conv. W						
Mean	55.86	31.52	34.79	1.21	21.54	29.42
Minimum	22.70	7.70	15.50	0.30	8.30	5.20
Maximum	113.60	68.40	80.30	2.50	37.80	68.00
Satira						
Mean	50.02	32.64	16.92	1.04	18.40	23.68
Minimum	45.90	29.90	15.70	0.90	14.60	16.90
Maximum	53.70	35.30	18.30	1.30	20.90	29.20
Ouzra						
Mean	52.54	33.94	18.54	1.06	19.38	26.14
Minimum	50.90	33.10	16.80	1.00	18.00	22.40
Maximum	55.70	36.10	27.50	1.20	21.10	28.90
Irrisaala						
Mean	63.28	39.18	12.77	1.27	21.53	29.73
Minimum	57.90	29.20	8.90	1.20	19.90	28.90
Maximum	68.70	49.80	14.90	1.40	22.50	30.20

Spatial distribution of different metals show that the industrial zone is the most polluted zone where you have the highest average of total metals content (300.9 mg.kg-1). Globally, the sum of these concentrations decreased gradually with increasing distance from the industrial area to the south (Khlédia Zone).

To see more the possible contribution part of industrial parks and TWW to the soil pollution by heavy metal metals, we draw four radials (Fig. 1): A, B, C, D (Fig. 3). Their origins are located in industrials area and near the coast of Tunis Gulf and its lower limits are in the South part of the plain. In addition the B, C and D radials pass respectively by Sadira, Ouezra and Irrisaala irrigated perimeters.

Table 3: sample numbers used for each radial

	1	**2**	**3**	**4**	**5**	**6**	**7**	**8**
A		31	29	13	15	5	4	1
B	30	34	27	7	21	40	24	6
C			8	28	14	39	41	19
D			12	16	22	10	11	17

To can easily read the figure 3, we have proposed the table 3. The numbers 1, 2, 3 ... correspond to abscissa points. A, B, C, D are the radial plotted in Figures 1 and 3. The rest of the numbers correspond to samples numbers. For example, the point 2 of abscissa axis of the radial A figure 3 is the sample N° 31 (based on table 3) of figure 2. The sum of HM concentration of sample 31 is 299.4 mg kg-1 (Table 4).

Table 4: the level of different heavy metals concentrations
For the 4 radials

	1	**2**	**3**	**4**	**5**	**6**	**7**	**8**
A		299,4	275,4	252,9	187,6	103,5	99,8	104,5
B	330,4	308,5	214,1	146,8	108,9	142,1	139,9	107,2
C			340,6	206,4	114,1	162,1	153,3	118,4
D			300,2	227,5	143,4	176,9	166,1	102,3

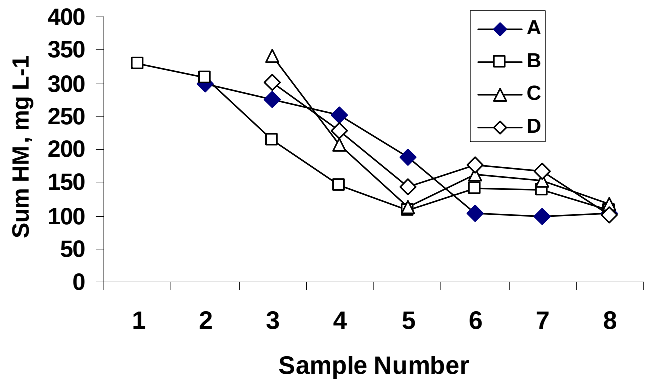

Fig. 3: The sum of HM concentration of samples along the for radials

The figure 3 shows that the metal concentration of samples from the industrial parks soil (points 1 and 2, fig. 3) are variables but it are the most polluted. When we move towards the southern part, the metal concentrations continue to be variable but for the same radial always decrease (points 3, 4 and 5, Fig.3). If the radial does not pass through an irrigated perimeter by TWW, the decline continues (points 6, 7 and 8; radial A in Fig. 3). But if we pass by an irrigated soil with TWW (points 6 and 7; radial B, C and D in Fig. 3) soils show a slight and significant increase of heavy metals levels. We also note that the average metals concentrations of Irrisaala is slightly higher (mean = 167.8 mg.kg-1) than in Sadira and Ouezra were the averages sum are respectively 141.8 mg.kg -1 and 148.2 mg.kg-1. It is noted that the last two averages difference is not very important but you took into account this variation because it is found that some trace elements (metals) concentrations show relatively an important variations from one sector to other

. These results are in agreement with previous investigations (Bahri, 1987; Mazlan et al., 1994; Clay et al., 2009) showing that irrigation by treated wastewater increases the content of heavy metals in soil surface horizons.

Later on, the metals levels regression takes again (point 8, Fig. 3). These results show that the industrial zone is a major pollution source. Pollutants are, in part, discharged directly on site. Aeolian transport might play a significant role, especially at short distance from the source. Moreover, the used treated wastewater contributes to soil pollution.

But you must never neglect that the spatial differences in metals can be due to variable heavy metal leaching towards the deep horizons.

We only discussed the sum of HM analyzed. So initially, we try to know if all analyzed metals increased or decreased at the same time and what are the factors that determine retention or percolation of these pollutants.

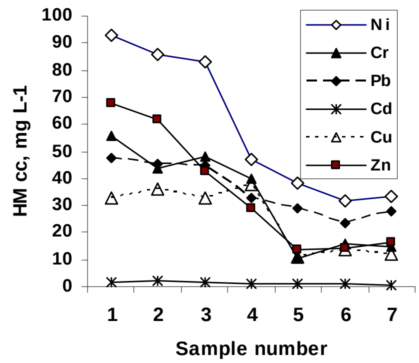

Fig. 4: Metal content variations along Radial A

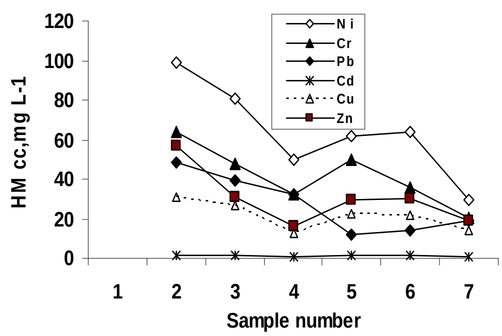

Fig. 5: Metal content variations along Radial D

We tested the tenors variations of Ni, Cr, Pb, Cd, Cu and Zn (series 1-6) along the for radials. For example, the concentration of each metal for each sample and for the same radial (A and D radials given as example; Figs. 4 and 5) are variable and all metals does not follow the same trend change. These variations may be due to different metal retention which is governed probably by soil texture (Tam and Wong, 1999) or/and soil composition, especially the organic matter levels (Clay et al., 2009) due to their high adsorption and absorption capacity. Also, the mobility of metals is closely related to soil physicochemical parameters such as soil pH and salinity and the global quality of the irrigation water.

4.2. Heavy metals and Soil constitution (organic matter and clay)

Heavy metal mobilization depends on the variability of soil structural texture and composition (Mapanda et al., 2005, Serpaud et al., 1994). Good correlations are well documented in the literature between heavy metals concentrations, mineralogical characteristics and soil texture. These parameters can be important variables which play a non negligible role in the metals retention.

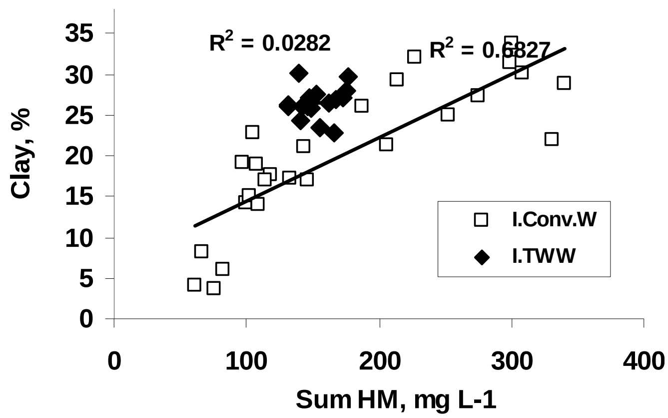

Fig. 6: correlation diagram showing total metal content versus per cent clay

Figure 6 shows that in irrigated perimeters by TWW the clay contents is not decisive in the metals retention because the correlation coefficient is only 0.02. However, the metals tenors in soils irrigated by water from the aquifers and Mejreda-Cap Bon channel are widely correlated to clay fraction. They also depend on other parameters because R^2 is only 0.68.

The organic matter currently considered as the most important reservoir of nutritive substances for soil. The organic matter associated to the solid phases can play too an important role on metal immobilization (Janos et al. 2004). The heavy metals can be adsorbed by organic matter in soil and/or form insoluble organometallic complexes (Sauvé et al., 2000; Udom et al., 2004). Correlation diagram shows that relatively abundant organic matter of the soil irrigated by TWW retains heavy metals since the correlation coefficient is 0.52. This retention is probably due to adsorption processes. But it is possible that the pH increase can provoke the metals precipitation. Also and following the organic compounds evolution, we have formation with humic substances of humic-heavy metal stable complexes (Pigizzo, et al., 2006) and increasing of metal stability.

While for soils irrigated by conventional water where TOC percentage are often lower than those of Naassen and Irrisaala, the immobilized metals quantity is variable and no relationship exists between organic compound yields and the total metals concentration (correlation coefficient close to 0). This result indicated that clay and organic matter associated to soil matrix do not play always together an important role on metal immobilization as proposed by many authors (eg, Janos et al., 2004; Clay et al., 2009) but the retention mechanism of metals also depends on the history of each irrigation district.

So, the heavy metal mobility must depends their chemical speciation, too, witch is closely related to water quality used for irrigation and soil chemical parameters as pH. To test the differentiation origin of the likely heavy metals mobility analyzed in our studied area, we adopt the sequential extraction technique.

4. Conclusion

The soil pH varies between 6.8 and 8.4. The TOC and E.C. are respectively between 0.10% and 1.40% and between 1.1 and 1.57 mS/cm. The soil Irrisaala has the highest pH, E.C. and TOC. While at Khlédia we have the weakest pH, E.C. and TOC of Mornag plain. Given the physicochemical characteristics of different irrigation water, organic compounds added to soil by TWW have caused a change in pH, TOC. and E.C. of these soil. The average concentrations of heavy metals in surface sediments range from 1.18 to 113.6 mg.kg-1. The heavy metal abundance order is Ni> Cr> Zn> Pb> Cu> Cd. these results show that industrial zone is a major pollution source. Treated wastewater irrigation also contributes to surface soil pollution. The concentrations of each metal in each sample and for the same area are variable and all metals do not follow the same variation trend. The retention of heavy metals is favored by the soil organic fraction in irrigated perimeters by TWW. However, in the rest of the areas the clay fraction of the solid matrix adsorbed a non negligible quantity of metal pollutants.

Heavy metals in soil fractions have similar distribution in the same sector which have the same history but differ for the same metal kind from one sector to other. In irrigated perimeters by TWW and the Artificial Refill zone, the water pH is an important factor that determines the metal mobility.

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