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Environmental Risks Associated with The Leakage of Untreated Wastewaters in Industrial Areas

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MANY countries around the world are forced to use wastewaters in plant production and their codes regulate such usage. However, these policies are not enough to stop their indirect impacts on the ecosystem. In Shobra El-Khema (Egypt), canals and agricultural drains still receive massive amounts of untreated industrial wastes and; therefore, the Egyptian government prohibited the usage of this wastewater for crop production. Alternatively, farmers use well water for irrigation; however, these shallow wells still receive wastewater leakage from the main drains and this probably possesses negative implications on the environment immediately or at least in the long-term. Accordingly, water, soil and plant samples were collected from these wastewater - contaminated areas wherein their contents of Fe, Mn, Zn, Cu, Cd, Co, Ni and Pb were investigated. The comprehensive pollution index (CPI) was then calculated for potentially toxic elements (PTEs) in water of both the main drain and the well waters (during the winter and summer seasons of 2016, 2017 and 2018). Results indicate that there was no significant variation in CPI between these two sources ($P<0.05$). On the other hand, ammonium bicarbonate–diethylene triamine penta acetic acid (AB-DTPA) extractable PTEs and their total contents increased progressively and significantly within the subsequent soil layers during the three successive years of study ($P<0.05$). In this concern, the highest concentrations of PTEs were detected within the top (0-15 cm) soil layer, while decreased with increasing soil depth. The grown plants nearby the main drain (especially leafy vegetables) accumulated high concentrations of PTEs in their shoots and these concentrations exceed the permissible levels of Pb and Cd. On the other hand, levels of soil pollution with PTEs decreased considerably with increasing the distance from the main drain in spite of that slight to moderate levels of soil pollution with PTEs were detected at 1000 m apart from the main drain. Thus, leakage of untreated wastewaters possesses high ecological risks that extents to a distance of one kilometer apart from the main drain.

Keywords: Untreated wastewater; Environmental risks; Shallow wells; Potentially toxic elements; Soil pollution index; Comprehensive pollution index; Organic pollution index.

Introduction

Water scarcity has become the major concern that faces crop production in many arid and semi-arid countries (Abbas and Bassouny, 2018, Fitton et al., 2019 and Pereira et al., 2019). These countries were; therefore, forced towards using wastewaters in crop production as a socio-economic necessity to substitute partially fresh water insufficiency (Mapanda et al., 2005). However, these waters might, on the other hand, bring up elevated concentrations of potentially toxic elements (PTEs) to soils (Mapanda et al., 2005; ElShazly et al., 2019) beside of pathogens (Elbana et al., 2017) that might enclose potentially health risks to human and the ecosystem (Abdelhafez et al., 2015 and Abuzaid et al., 2019). Thus, regular monitoring of contaminants in effluents and sewage waters is essential to ensure their safe use in crop production

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(Arora et al., 2008). It is worthy to mention that the governmental bodies around the world have considered treating these wastewaters properly to ensure their safe use in plant production. In Egypt, the "Ministry of Land Reclamation and Agriculture, Ministry of Water Resources and Irrigation, Ministry of the Environment, Egyptian Environmental Affairs Agency, Ministry of Housing Utilities and Urban Communities, and Ministry of Health and Population" are concerned with the reuse of treated wastewater (TWW) in Agriculture (Elbana et al., 2017). Although, these bodies regulate and manage the reuse of TWW in crop production; however, these regulation did not consider the indirect impacts of these waters on the surrounding arable lands; especially when hydraulic continuity exists between wastewaters in drains and the well waters of these arable lands as highlighted by Farid et al. (2019).

Other anthropogenic activities, *e.g.* smelting (Mohamed et al., 2018), aerosols precipitation from the nearby industrial areas (Shaltout et al., 2014) and/or heavy traffic highways (Hashim et al., 2017) can bring considerable concentrations of PTEs to soils. These contaminants are non bio-degradable and consequently persist in soils for years (Azimi et al., 2017). Plants grown on such contaminated areas absorb considerable concentrations of these contaminants especially vegetables (Jolly et al., 2013). Thus, PTEs enter the food chain (Abdelhafez et al., 2015) and cause a "number of nervous, cardiovascular, renal, neurological impairment as well as bone diseases and several other health disorders" (Jolly et al., 2013). Accordingly, levels of PTEs determine food and feed safety (Tóth et al., 2016).

The arable lands nearby the industrial areas may suffer severely from soil pollution especially with PTEs. In this concern, Shobra El-Khema soils are considered the ideal models to investigate the consequences of the industrial revolution on PTEs pollution in the arid zone soils. This area is located at the southern part of the Nile Delta in Egypt. Its arable lands receives atmospheric aerosols from the nearby industrial areas (Safar & Labib, 2010 and Shaltout et al., 2014). Moreover, its open agricultural drains receive massive amounts of the industrial wastes (Abdel-Shafy and Aly, 2007). This water has been partially used to substitute fresh water in crop production, however, the consequences of using this water signal pollution hazards (Melegy and Chin, 2004). Accordingly, the Egyptian government prohibited the usage of this wastewater for irrigation without offering suitable alternative sources for irrigation. The farmers were, therefore, forced to use waters of underground shallow dug wells for irrigation. It is thought that waters of these shallow wells still receive wastewater leakage (indirect source of soil pollution) from the main (agricultural+ industrial) drains (hypothesis 1, H1). This non-point source of pollution is supposed to raise concentrations of PTEs in soil up to ones exceed their permissible levels (hypothesis 2, H2). Such soil contaminants might find their way to deeper soil layers (hypothesis 3, H3) and probably concentrate within the aerial parts of the grown plants to levels exceeding the permissible ones (hypothesis 4, H4). On the other hand, the levels of soil pollution with PTEs are expected to increase with decreasing distance between soil and source of pollution, *i.e.* the wastewater drain (hypothesis 5, H5).

Materials and Methods

Water, soil and plant samples were collected from the contaminated areas (the arable lands nearby the main drain) to determine their total and AB-DTPA extractable contents of PTEs in order to examine the hypotheses from 1 to 4 (H1, H2, H3 and H4). Further soil and plant samples were collected at 250, 500, 750 and 1000 m apart from the studied area (nearby the main drain) to investigate their total contents of PTEs with the aim of testing H5.

Site description

The studied area represents the arable lands nearby the industrial zone of the Second Shobra El-Khima District, Qalubia Governorate (North sector of Greater Cairo, Egypt). This area is located in the arid zone (total annual rainfall is 5 mm) with an average temperature of 27.5°C in summer and 13.3°C in winter. The wind speed is 12 km h-1 and generally blows from the north and north east directions. This area encompasses a strange-mixture of anthropogenic activities (*e.g.* housing, agricultural and industrial ones).

Soil, plant and water sampling

Soil samples were collected at different depths *i.e.,* 0 to 15, 15 to 30, 30 to 60 and 60 to 120 cm from the arable lands nearby the main drain during three successive years i.e. 2016, 2017 and 2018. A reference soil was selected for data comparison (Fig. 1). Three more locations were considered for soil sampling during 2018 only (250 m apart from each other on the opposite side of Shibin

El-Qanater collector drain). In this concern, three sites were selected to represent the replicates of each point of the study.

These samples were air dried, crushed, sieved to pass through 2 mm sieve and then analyzed for their chemical properties as outlined by Sparks et al. (1996). Chemical characteristics of the investigated soils are presented in Table 1.

Physical characteristics of the investigated soil and the background one were conducted according to Klute (1986) and the results are presented in Table 2.

Also, irrigation water (shallow wells) and plant shoot *i.e.,* spinach (Spinacia oleracea), molokhia (Corchorus olitorius), wheat (Triticum aestivum), maize (Zea mays) and cabbage (Brassica oleracea var. capitata) were sampled from the investigated areas during the abovementioned periods. Moreover, water samples were collected from Shibin El-Qanater collector drain that receives wastewater from plastics- dyeing and textile - steel - paper and detergent factories, sewage effluents and domestic wastes nearby each investigated area during the sampling periods.

Fig. 1. Location map of the studied sites (Sites 1, 2 and 3 are the sites nearby the drain, sites 4, 5 and 6 represent the location at 250 m apart from the drain. Sites 7, 8 and 9 stand for the location at 500 m apart from the drain. Sites 10, 11 and 12 indicate the location at 750 m apart from the drain. Sites 13, 14 and 15 denote the location at 1000 m apart from the drain. Sites 16, 17 and 18 represent the background soil)

TABLE 1. Chemical characteristics of the investigated soil (nearby the drain) during 2016, 2017 and 2018 as well as the characteristics of the background one

Soil depth (cm)	pH*	EC**	CEC	$CaCO_3$	Gypsum	SOM
		dS m^{-1}	$cmol_c$ kg^{-1}	g kg^{-1}	g kg^{-1}	g kg^{-1}
			2016			
0-15	7.21	1.49	36.3	48	13.2	21.9
15-30	6.92	0.95	35.6	41	12.1	18.6
30-60	7.01	0.61	34.1	38.7	10.1	11.2
60-120	6.53	0.49	33.8	27.5	8.9	4.5
			2017			
0-15	7.28	1.54	39.2	50.1	14.5	22.1
15-30	7.02	1.02	37.4	42.2	13.3	19.1
30-60	7.12	0.69	36.6	39.4	11.2	11.8
60-120	6.76	0.57	34.9	28.9	9.5	6.3
			2018			
0-15	7.34	1.59	41.1	51.2	16.2	22.6
15-30	7.11	1.08	39.3	43.4	14.5	19.4
30-60	7.14	0.73	37.9	39.8	11.2	12.1
60-120	6.82	0.61	36.2	29.1	9.5	7.6
			Background soil			
0-15	7.11	1.23	34.5	42	12.4	21.1
15-30	6.74	0.72	33.7	38.5	11.7	16.3
30-60	6.89	0.45	32.8	35.4	9.4	10.6
60-120	6.41	0.28	31.3	26.2	6.7	3.4

Note:. pH*: in 1:2.5 soil: water suspension; EC**: in saturated paste extract; CEC is the cation exchange capacity and SOM is the soil organic matter

TABLE 2. Physical characteristics of the investigated soil (nearby the drain) and the background one

	Soil nearby the drain						Background soil					
Soil depth	Sand %	Silt %	Clay %	Texture (USDA)	HC cm h^{-1}	BD Mgm^3	Sand %	Silt %	Clay %	Texture (USDA)	HC cm h^{-1}	BD Mgm^3
0-15 cm	10.58	26.69	62.73	Clay	0.61	1.17	10.6	27.65	61.75	Clay	0.55	1.24
15-30 cm	11.55	25.84	62.61	Clay	0.56	1.19	11.61	26.73	61.66	Clay	0.53	1.26
30-60 cm	13.16	24.74	62.1	Clay	0.53	1.21	13.22	25.71	61.07	Clay	0.52	1.28
60-120cm	13.76	26.65	59.59	Clay	0.51	1.24	13.89	27.6	58.51	Clay	0.49	1.29

Note: HC is the saturated hydraulic conductivity and BD is the bulk density.

Water, soil and plant analyses

Water samples were characterized for their chemical properties as outlined by Nollet and De Gelder (2014) and the results are demonstrated in Table 3. Chemical oxygen demand (COD) and biological oxygen demand (BOD) in both the main drain and the underground water (that is used for irrigation) were determined according to Young (1973).

Ammonium bicarbonate–diethylene triamine penta acetic acid(AB-DTPA) concentrations of PTEs were extracted according to Soltanpour (1985). Soil portions (< 2mm, equivalent to 0.5 g on dry basis) were acid digested using a tri-acid mixture (HNO_3:H_2SO_4:$HClO_4$, 10:4:1) as mentioned by Sahrawat et al. (2002). The collected plant samples were cut into small pieces; oven dried at 105° C for 72 h, and then acid digested using a mixture of concentrated sulphuric acid and perchloric acid (2:1) according to Piper (2019)

Total concentrations of PTEs in the water samples, soil and plant digests as well as the AB-DTPA soil extracts were measured by Inductively Coupled Plasma Optical Emission Spectrometer (ICP-OES) Model PERKIN ELMER Optima 3000.All the chemicals used in this study were of analytical grade reagent.

TABLE 3.Chemical characteristics of the well water and the water of the main drain

	Water of the main drain						Well water					
	2016		2017		2018		2016		2017		2018	
Properties	**Winter**	**Summer**	**Winter**	**Summer**	**Winter**	**Summer**	**Winter**	**Summer**	**Winter**	**Summer**	**Winter**	**Summer**
pH	8.30	8.10	8.50	8.30	8.80	8.60	8.64	8.78	8.59	8.74	8.55	8.72
EC, dS/m	1.88	1.81	1.94	1.87	2.05	1.95	2.20	2.75	2.17	2.71	2.12	2.67
TDS, mg L^{-1}	1203	1154	1241	1196	1312	1248	1408	1760	1388	1734	1356	1708
TH, mg L^{-1}	410	379	429	388	441	399	451	569	446	565	443	561
SAR	7.65	7.11	7.74	7.17	7.81	7.25	7.49	7.98	7.45	7.94	7.42	7.91
Mg ratio ,%	50.20	49.80	52.60	50.90	55.80	51.10	52.90	55.40	52.20	54.90	51.70	54.10
NO_3^- , mg L^{-1}	0.82	0.94	0.89	0.99	0.94	1.03	1.96	1.85	1.88	1.7	1.81	1.60
NH_4^+ , mg L^{-1}	3.87	4.43	3.98	4.47	4.21	4.79	4.94	4.67	4.87	4.56	4.82	4.50
PO4 , mg L^{-1}	1.07	1.41	1.11	1.55	1.34	1.83	1.99	1.64	1.94	1.59	1.90	1.53
DOC, mg C L^{-1}	167	178	172	183	185	197	195	176	188	169	181	163
Fe , mg L^{-1}	1.60	1.67	1.69	1.76	1.82	1.91	2.16	2.08	2.09	1.97	2.04	1.92
Mn , mg L^{-1}	0.62	0.68	0.64	0.68	0.67	0.70	0.71	0.68	0.71	0.68	0.70	0.67
Cu, mg L^{-1}	0.14	0.11	0.18	0.14	0.25	0.19	0.16	0.19	0.14	0.17	0.11	0.15
Zn , mg L^{-1}	1.78	1.59	1.86	1.68	1.95	1.78	1.58	1.75	1.47	1.69	1.43	1.61
Cd , mg L^{-1}	0.01	0.02	0.03	0.04	0.06	0.08	0.06	0.04	0.04	0.02	0.02	0.01
Co , mg L^{-1}	0.18	0.13	0.21	0.17	0.28	0.21	0.21	0.29	0.18	0.24	0.16	0.21
Pb , mg L^{-1}	0.36	0.43	0.43	0.50	0.56	0.69	0.53	0.40	0.49	0.35	0.46	0.31
Ni, mg L^{-1}	0.17	0.24	0.23	0.29	0.31	0.42	0.38	0.31	0.31	0.26	0.27	0.21

Note: EC is the electric conductivity, TDS is the total dissolved solids, TH is the total hardness, SAR is the sodium adsorption ration and DOC is the dissolved organic carbon.

Pollution indices and statistics analysis

Soil pollution index was calculated as relations between the concentrations of PTEs in soil and the corresponding background ones (in the reference soil).

$$\text{Soil pollution index}^{(SPI)} = \frac{\text{concentrations of PTE in soil}}{\text{Background concentrations of PTEs}} \quad (1)$$

This index was compared with the intervals of the pollution indexes presented by Lacatusu (2000) *i.e.,* values from 1.1-2.0 (slight pollution), 2.1-4.0 (moderate pollution), 4.1-8.0 (severe pollution), 8.1 - 16.0 (very severe pollution), > 16.0 (excessive pollution).Comprehensive pollution index (CPI) with PTE and the organic pollution index (OPI) were calculated in the water samples according to Shakir et al. (2017) as follows:

$$\text{Comprehensive pollution index CPI} = \frac{1}{n}\Sigma \text{Water pollution index} \quad (2)$$

where n is the number of parameters

$$\text{Water pollution index}^{(WPI)} = \frac{\text{(Concentration of PTEs in water)}}{\text{Their permissible levels}} \quad (3)$$

According to the calculated values of CPI, water quality can be evaluated into: clean: (index ≤ 0.2), sub clean (index ranged from 0.21 to 0.4), slightly polluted (index ranged from 0.41 to 1.0), moderately polluted: (index ranged from 1.01 to 2.0), and severely polluted: (index ≥ 2.01)

$$\text{Organic pollution index}^{(OPI)} = \frac{BOD}{BOD_{ref}} + \frac{COD}{COD_{ref}} + \frac{\text{Nitrate}}{\text{Nitrate}_{ref}} + \frac{\text{phosphate}}{\text{phosphate}_{ref}} \quad (4)$$

The obtained data were then statistically analyzed using PASW Statistics software 18.

Results and Discussion

Water pollution indexes for evaluating the quality of the well water and the water of the main drain

Concentrations of Mn, Cd, Co and Ni in shallow wells (collected from the investigated area) exceeded the limits allowed by ECP 501 (2015) in treated irrigation water i.e.: 0.2 (Mn), 0.01 (Cd), 0.05 (Co) and 0.2 (Ni) mg L^{-1}. On the other hand, concentrations of Fe, Cu, Zn and Pb were below the permissible levels (i.e. 5 (Fe), 0.2 (Cu), 5 (Zn) and 5 (Pb) mg L^{-1}) in shallow wells. Such results indicate the potential hazards of using such water for irrigation.

Comprehensive pollution index (CPI) with PTEs

Pollution indexes (PI) were calculated, for waters of both the main drain and the source of irrigation (the shallow wells), to evaluate the potential risks of PTEs in both water resources. Afterwards, the comprehensive pollution index (CPI) was calculated and the obtained data are presented in Figure 2. It seems that the investigated waters were moderately polluted with PTEs (CPI<2) during 2016 and 2017 as outlined by Shakir et al. (2017). However, these waters became severely polluted with PTEs (CPI<2) during 2018. Data analysis reveal that there were no significant variations in CPI values between these two sources of water according to “T-test” in pair (T=0.633, *P*=0.555).

For more details, regression studies were conducted between concentrations of PTEs in the well water and the corresponding ones of the main drain (Fig. 3).

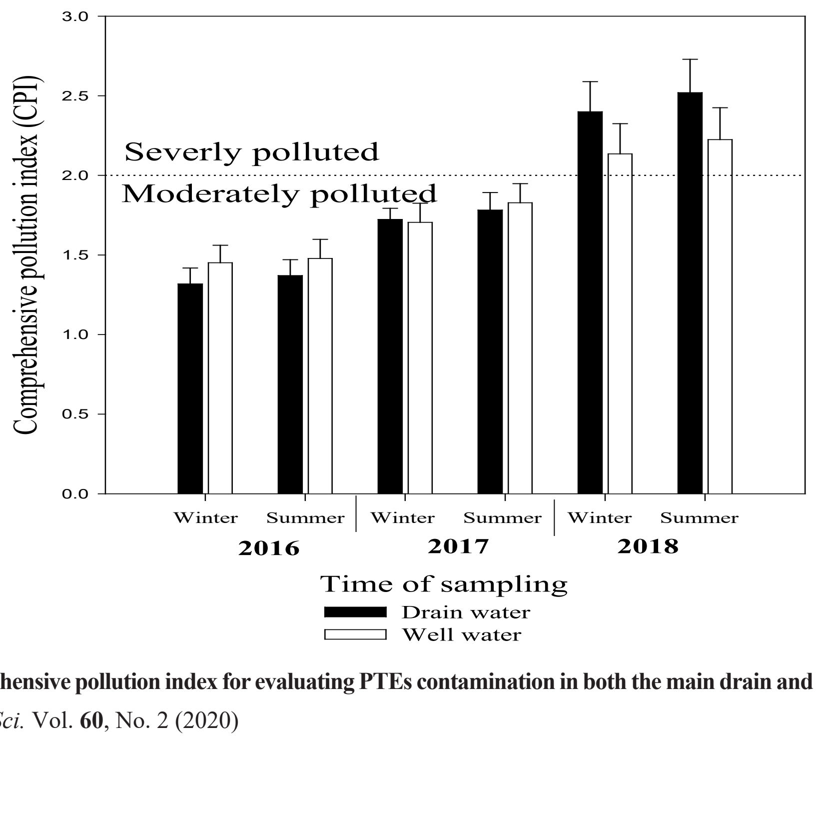

Fig. 2. Comprehensive pollution index for evaluating PTEs contamination in both the main drain and underground waters

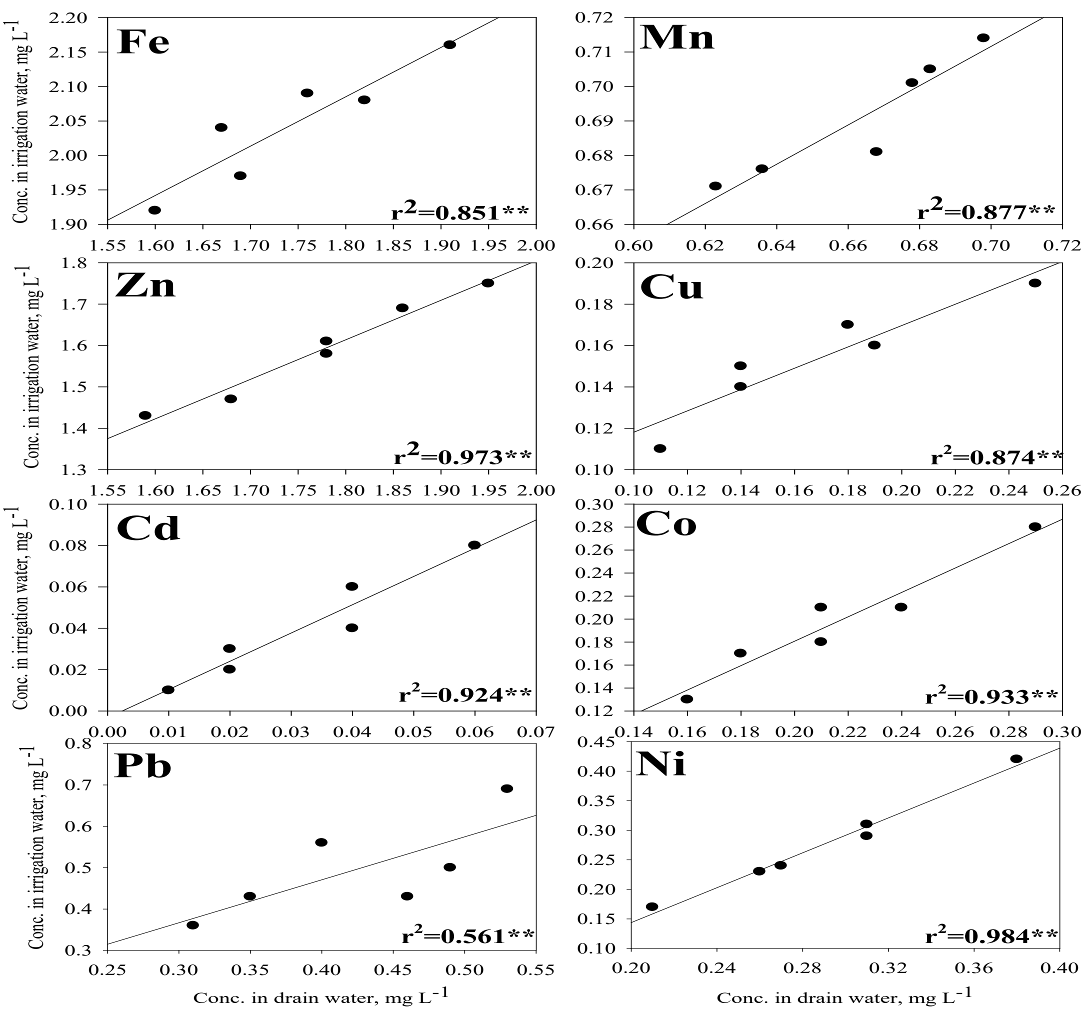

Fig. 3. Regression relations between concentrations of PTEs in the well water and the corresponding ones in the main drain (every point is a mean value of three replicates collected at both the summer and winter seasons of years 2016, 2017 and 2018)

Organic pollution index (OPI)

Organic pollution was also considered while evaluating the suitability of the investigated waters for irrigation. To attain this goal, the following indicators, recommended by Shakir et al. (2016), were taken into account i.e. chemical oxygen demand (COD), biological oxygen demand (BOD) and organic pollution index (OPI). The biological oxygen demand (BOD) expresses the potentiality of removing oxygen from wastewater by aerobic heterotrophic bacteria (Bourgeois et al., 2001) through respiration or catabolism (Horan, 2003) within a period of five days (Scholz, 2006). Its acceptable level is thought to be 30 mg L^{-1} or less (Salameh et al., 2018; Singh et al., 2019). Shockingly, the obtained values for both the irrigation well water and the drain water were 7.5-9 folds higher than the acceptable levels (Fig 4). Probably, such high values indicate potential eutrophication and changes in the ecosystem (Oladoja et al., 2017). Another aspect that should be considered regarding organic pollution in water is the chemical oxygen demand (COD). This indicator measures the organic loads in wastewaters (Wichern et al., 2018) which micro-organisms cannot metabolize within five days period (Scholz, 2006) while measured using strong oxidizing agent under acid conditions (Bourgeois et al., 2001). Its acceptable value should not exceed 200 mg L^{-1} (Bhuyan et al., 2018). Unfortunately all studied water samples exceeded the permissible levels and went beyond the maximum permissible limits recommended for COD and BOD in the secondary treated wastewater i.e. 80 and 40 mg L^{-1} according to the Decree 44/2000, Article 15 of the Executive Regulation of Law 93/1962 (Elbana et al., 2017).

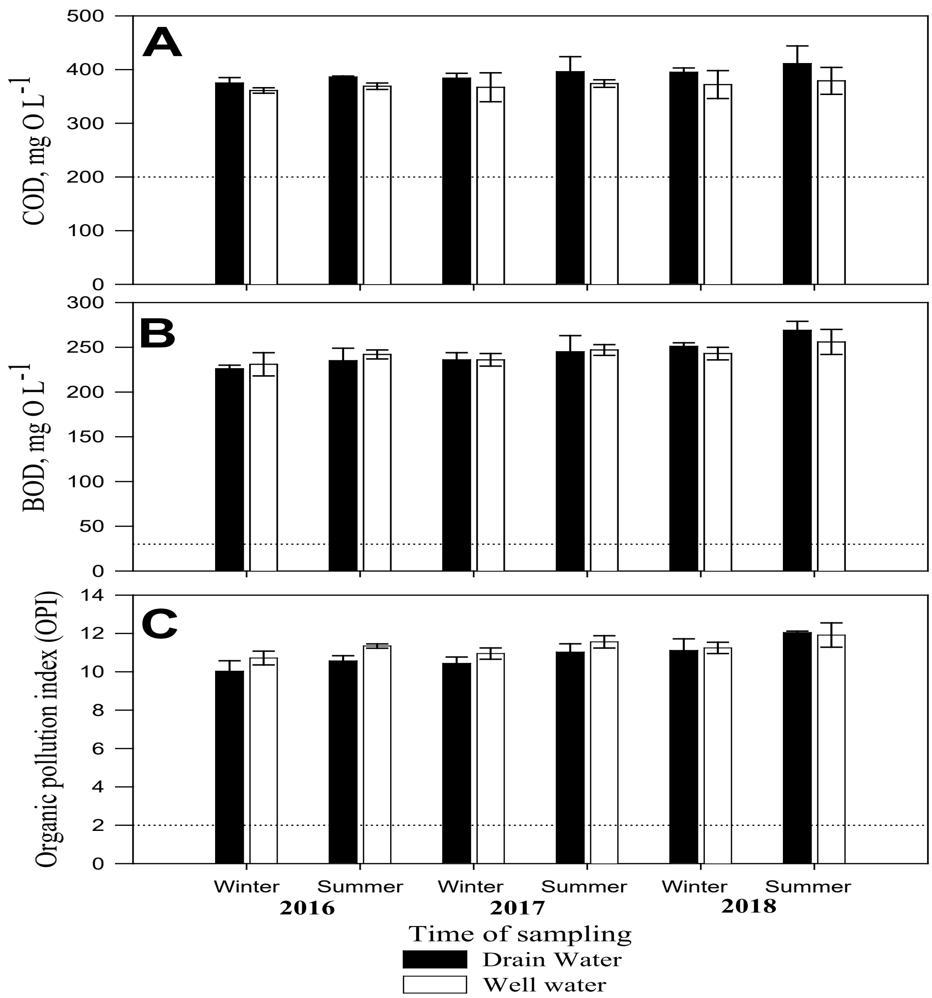

Fig. 4. Chemical oxygen demand (COD), biological oxygen demand (BOD) and organic pollution index (OPI) for evaluating PTEs contamination in both the main drain and well waters(the dot-lines of COD and BOD graphs represent their acceptable levels while the dot-line of OPI presents a level of severe pollution with PTEs)

Organic pollution index (OPI) is a combined meter that considers each of BOD, COD, nitrate, and phosphate concentrations in the evaluation process of water quality (Shakir et al., 2017). This index classifies wastewaters quality into different classes based on the level of organic pollution (Wei et al., 2009; Chaudhary et al., 2017). It seems that all the collected water samples were severely contaminated with organic pollutants (OPI values >2).

Based on the "T-test" results, values of BOD of the irrigation water were comparable with the corresponding ones of the drain water ($T_{n=18}$=0.546, *P*= 0.592); however, the other organic pollution indexes differed significantly between these two sources *i.e.,* OPI (T=0.205, *P*<0.001) and COD (T=8.440, *P*<0.001).This result might indicate that the organic pollution that occurs in the underground water might also be affected by the leakage of mineral and organic inputs for agrochemical from the top soil of the arable lands.

A correlation study was conducted to investigate the relationship between the organic pollution markers of irrigation water and the corresponding ones of the drain water (Fig 5). According to the calculated values of "r^2", the relationship between COD in the irrigation water and the corresponding ones in the underground water were highly significant. Thus, it can be deduced that hydraulic continuity probably exists between these two sources of irrigation water and therefore, we accept the first hypothesis, H1.

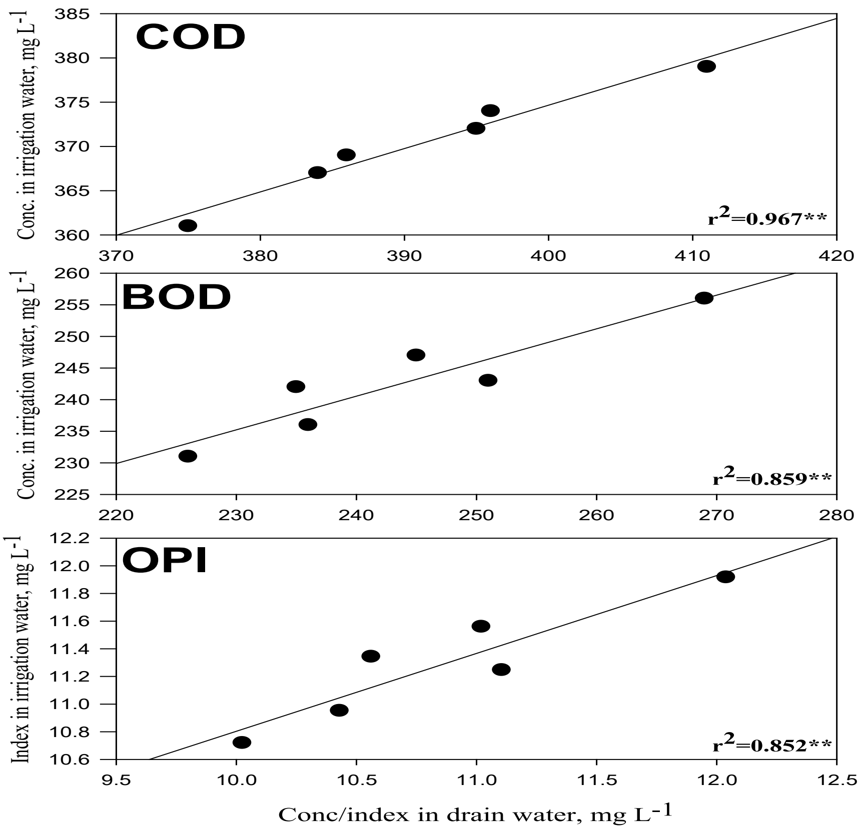

Fig. 5. Regression relations between values of chemical oxygen demand (COD), biological oxygen demand (BOD) and the organic pollution index (OPI) in the underground water and the corresponding ones in the main drain (every point is a mean value of three replicates collected at both the summer and winter seasons of years 2016, 2017 and 2018)

Implications of long term irrigation with wastewater on the total and AB-DTPA extractable concentrations of PTEs in soil

Figures 6 and 7 reveal that both AB-DTPA and total contents of Mn, Zn, Cu, Cd, Co and Ni increased progressively and significantly within the different soil layers during three successive years of study i.e. 2016, 2017 and 2018. To calculate the levels of soil pollution with PTEs, their total contents were determined in the reference soil at 60-120 cm soil depth, and those levels were selected to be the background ones. It was found that concentrations of PTEs in the contaminated area exceeded the background levels within the different soil layers. The corresponding increases that occurred in the top “0-30 cm” soil layer (during 2018) were 1.12 (Fe), 1.39 (Mn), 1.70 (Zn), 1.36 (Cu), 2.28 (Cd), 1.35 (Co), 1.28 (Pb) and 1.39 (Ni) fold higher than their reference levels. Probably, the elevated concentrations of PTEs in soil resulted from the concurrent increases that occurred in AB-DTPA PTEs in soil In this concern, the calculated correlation coefficients (r^2) between the AB-DTPA extractable PTEs and their corresponding total contents in soil were 0.847** (Mn), 0.979**.(Zn), 0.890** (Cu), 0.973**.(Cd), 0.912** (Co) and 0.949** (Ni), 0.849** (Pb).

Although, Fe and Pb concentrations increased noticeably within the different soil layers with ageing; however, such increases seemed to be insignificant. This might occur because the concentration of Fe in soil was probably high; and therefore, significant increase might only be expected after long term successive soil inputs. In case of Pb, other factors might contribute to soil pollution *e.g.,* aerosols precipitation (polluted with Pb) owing to heavy traffics (Hashim et al., 2017) and/or lead phosphate immobilization in the topsoil (Miretzky and Fernandez-Cirelli, 2008) due to the intensive fertilization with P-fertilizers (Melegy and Chin, 2004). Generally, concentrations of PTEs are in continuous increase and exceeded the permissible levels suggested by Kabata-Pendias (1995) and Khabata-Pendias and Pendias (2001) (1500 mg Mn kg^{-1}, 100 mg Pbkg^{-1}, 100 mg Ni kg^{-1}, 3 Cd mg kg^{-1}) within the surface and subsurface soil layers; however, in case of Co, its total content in soil did not exceed the permissible level (Co=50 mg kg^{-1}). Accordingly, we partially accept the second hypothesis, H2.

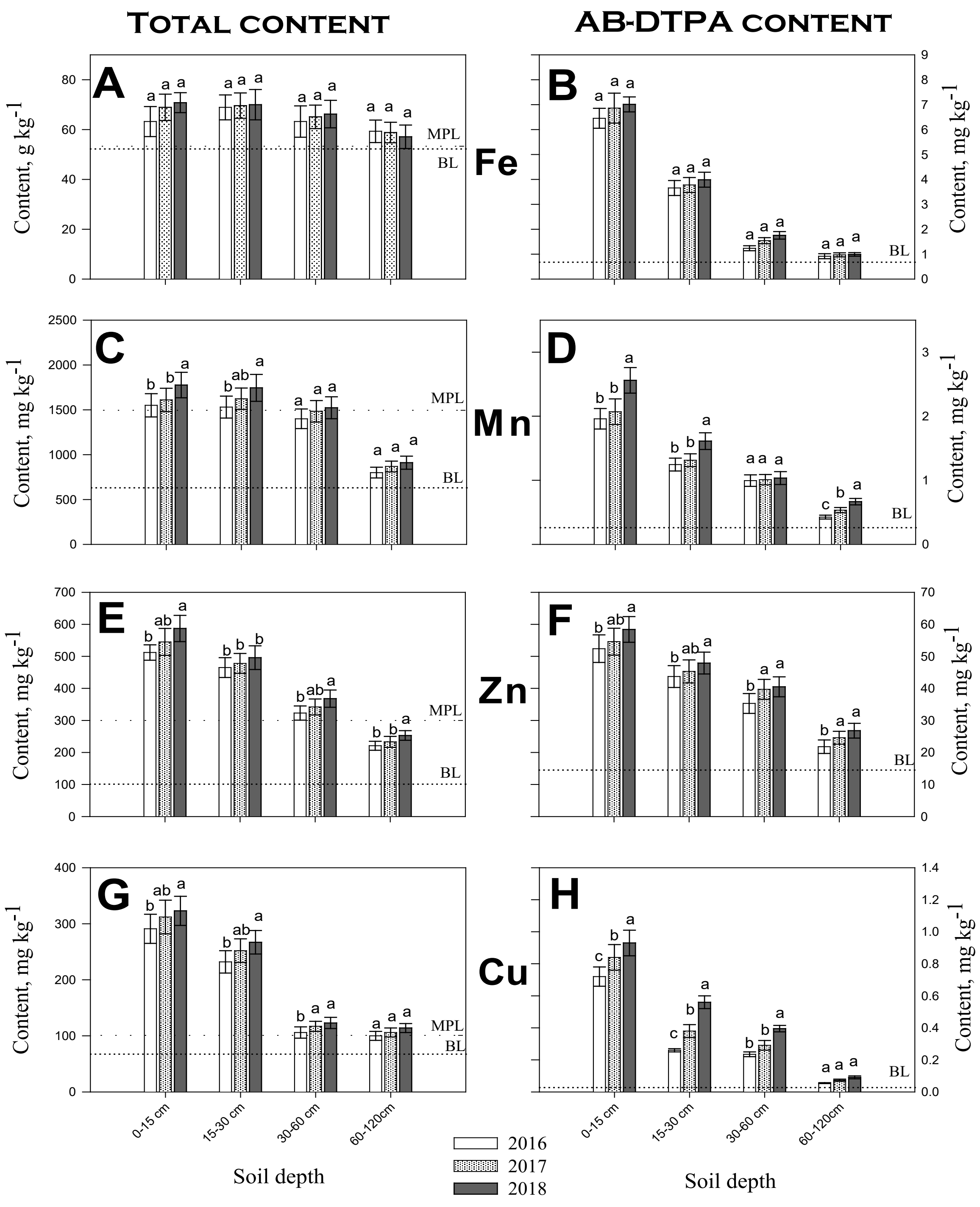

Fig. 6. Total and AB-DTPA extractable concentrations of Fe, Mn, Zn and Cu (mean ±SD) in the contaminated area nearby the drain ("BL" indicates the background level while "MPL" indicates the maximum permissible levels)

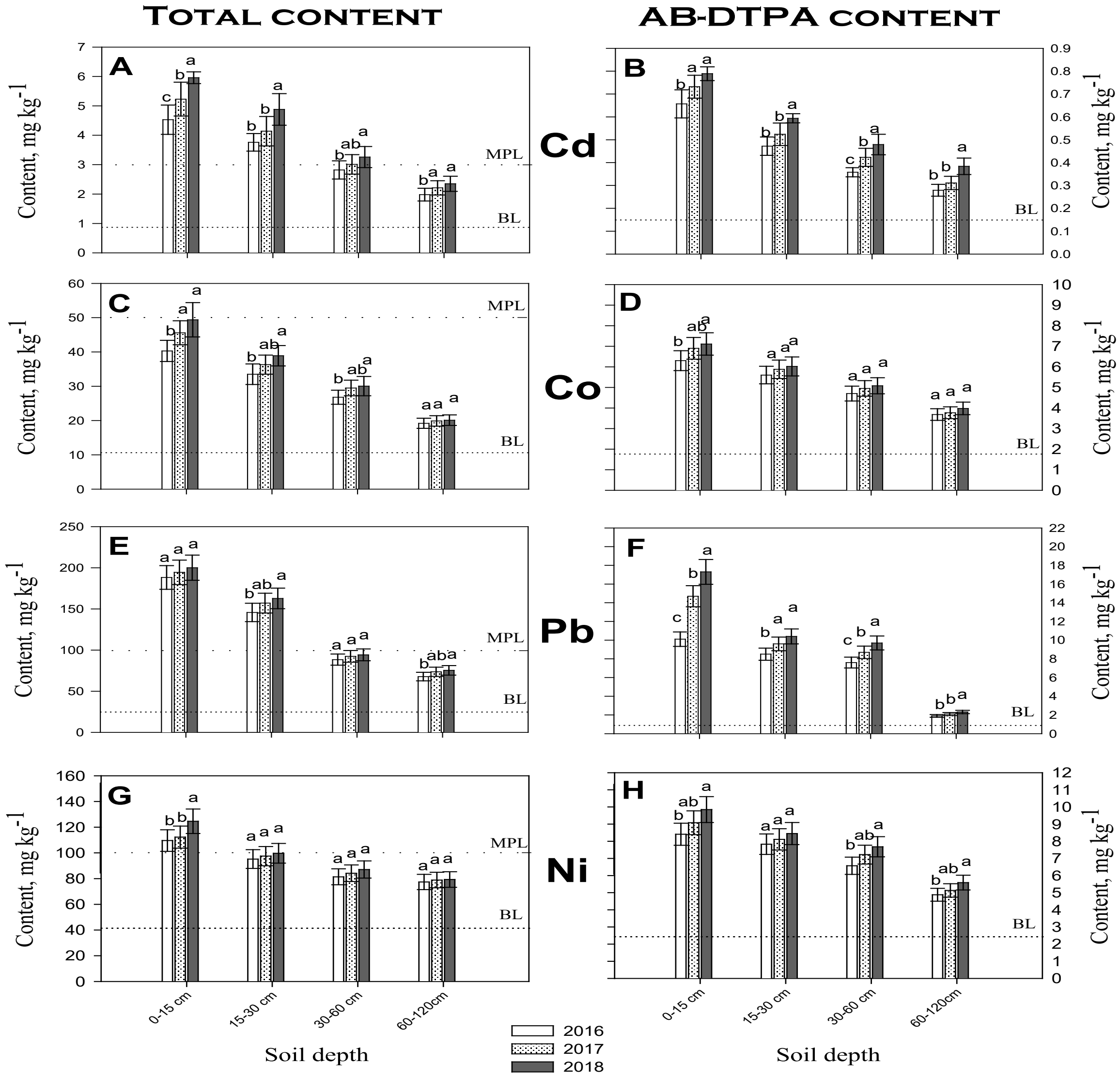

Fig 7. Total and AB-DTPA extractable concentrations of Cd, Co, Pb and Ni (mean ±SD) in the contaminated area nearby the drain ("BL" indicates the background level while "MPL" indicates the maximum permissible levels)

Distribution of PTEs within the different soil layers

The highest concentrations of PTEs (AB-DTPA extractable and total contents) were detected within the top (0-15 cm) soil layer while the corresponding concentrations seemed to be relatively lower within the subsequent soil layers (Fig. 6 and 7). It seems that PTEs concentrations were significantly and positively correlated with each of soil organic matter and CEC (Table 4). This might take place because the top soils received relatively higher amounts of SOM when compared with the subsequent soil layers (Abbas and Bassouny, 2018). This, in turn, probably improved considerably soil CEC; hence, increased the retention of PTEs in soil. Likewise, the total contents of PTEs correlated significantly with each of soil pH and $CaCO_3$ content. It is thought that, chemical equilibrium of PTEs in soil shifts toward the release of free metal cations at low pH values while precipitation dominates due to the formation of carbonate or hydroxyl complexes at high pH values (Rengel, 2015). Thus, precipitation might be a potential mechanism controlling PTEs behavior in the investigated soil (slightly alkaline). In this concern, the mobility of Fe, Mn and Zn decreased in soil with increasing soil pH (Rengel, 2015). Likewise, Co solubility decreased in soil with increasing soil pH (Smičiklas et al., 2015). Precipitation of PTEs as a function of soil pH can be illustrated according to the following equations adopted from Lindsay (1979) and Mishra et al. (2019):

$$\text{Soil-Fe}\,(Fe\,(OH)_3) + 3H^+ \leftrightarrows Fe^{3+} + 3H_2O \qquad (5)$$

$$\text{Soil - Zn} + 2H^+ \leftrightarrows Zn^{2+} \qquad (6)$$

$$\text{Soil - Cu} + 2H^+ \leftrightarrows Cu^{2+} \qquad (7)$$

$$\text{Soil - Cd} \leftrightarrows Cd^{2+} \qquad (8)$$

$$\text{Soil - Pb} \leftrightarrows Pb^{2+} \qquad (9)$$

Moreover, the hydrolysis of $CaCO_3$ probably increased the activity of OH^- ions consequently neutralized the activity of H^+ ions and shifted the above mentioned equations towards metal precipitation as outlined by **A**bbas and Salem (2011).

$$CaCO_3 + 2H_2O \leftrightarrow Ca^{2+} + 2OH^- + CO_2 \qquad (10)$$

Accordingly, we accept the third hypothesis, H3.

Concentrations of PTEs within shoots of plants grown on the contaminated area

Plants grown on the nearby polluted area accumulated high concentrations of PTEs in their shoots (Figs 8 and 9). This might occur because of the concurrent increases that occurred in both total and AB-DTPA extractable concentrations of PTEs in soil. In this concern, the leafy vegetables and Molokhia (*Corchorus olitorius*) accumulated relatively higher concentrations of PTEs than the field crops (wheat and maize) did. Moreover, concentrations of these contaminants in plant leaves were in continuous increases with increasing the cropping period. It seems that the concentrations obtained herein exceeded the sufficient or normal ranges adopted by Khabata-Pendias and Pendias (2001) and Kim et al. (2015) i.e. 1000 mg Fe kg^{-1}, 25-150 mg Zn kg^{-1}, 5-20 mg Cu kg^{-1}, 0.05-0.2 mg Cd kg^{-1}, 0.1-5 mg Ni kg^{-1}, 1-5 mg Pb kg^{-1}. Moreover, these concentrations were much higher than the corresponding ones detected by Mapanda et al. (2005) for leafy vegetables irrigated with wastewater in Zimbabwie for long time periods which ranged from 1.0 to 3.4 mg Cu kg^{-1}, 18 to 201 mg Zn kg^{-1}, 0.7 to 2.4 mg Cd kg^{-1}, 2.5 to 6.3 mg Ni kg^{-1} and 0.7 to 5.4 mg Pb kg^{-1}. Furthermore, concentrations of Cd and Pb obtained herein were higher than the permissible levels in edible leafy vegetables according to the recommendations of Joint FAO/WHO FOOD Standards (2011) *i.e.,* 0.2 mg Cd kg-1, 0.3 mg Pb kg^{-1}. In spite of that the green vegetables are widely grown in the investigated area and marketed among the population of this area. It is worthy to mention that the Egyptian Code prohibits the reuse of treated wastewater for production of any raw vegetables (Elbana et al., 2017). Since 1999, the level of Pb and Cd exceeded their permissible levels in 39-43% of the leafy vegetables in the Egyptian market (Dogheim et al., 2004) and our results support these findings which indicate that the measures of food safety are nearly absent in Egypt. Thus, we accept the fourth hypothesis, H4.

TABLE 4. Total PTEs contents in soil in relation with soil pH, SOM, CEC and $CaCO_3$

	pH	SOM	CEC	$CaCO_3$	Total-Fe	Total-Mn	Total-Zn	Total-Cu	Total-Ni	Total-Pb	Total-Co	Total-Cd
pH												
SOM	0.810**											
CEC	0.801**	0.647**										
$CaCO_3$	0.909**	0.946**	0.733**									
Total-Fe	0.137	0.249	0.342	0.177								
Total-Mn	0.891**	0.855**	0.782**	0.925**	0.356							
Total-Zn	0.848**	0.889**	0.864**	0.919**	0.289	0.906**						
Total-Cu	0.706**	0.941**	0.710**	0.878**	0.291	0.755**	0.910**					
Total-Ni	0.847**	0.827**	0.881**	0.867**	0.179	0.808**	0.963**	0.879**				
Total-Pb	0.767**	0.963**	0.722**	0.919**	0.253	0.799**	0.931**	0.991**	0.902**			
Total-Co	0.861**	0.944**	0.813**	0.961**	0.239	0.878**	0.958**	0.945**	0.932**	0.971**		
Total-Cd	0.819**	0.777**	0.919**	0.843**	0.315	0.839**	0.969**	0.852**	0.964**	0.864**	0.910**	

**. Correlation is significant at the 0.01 level (2-tailed).

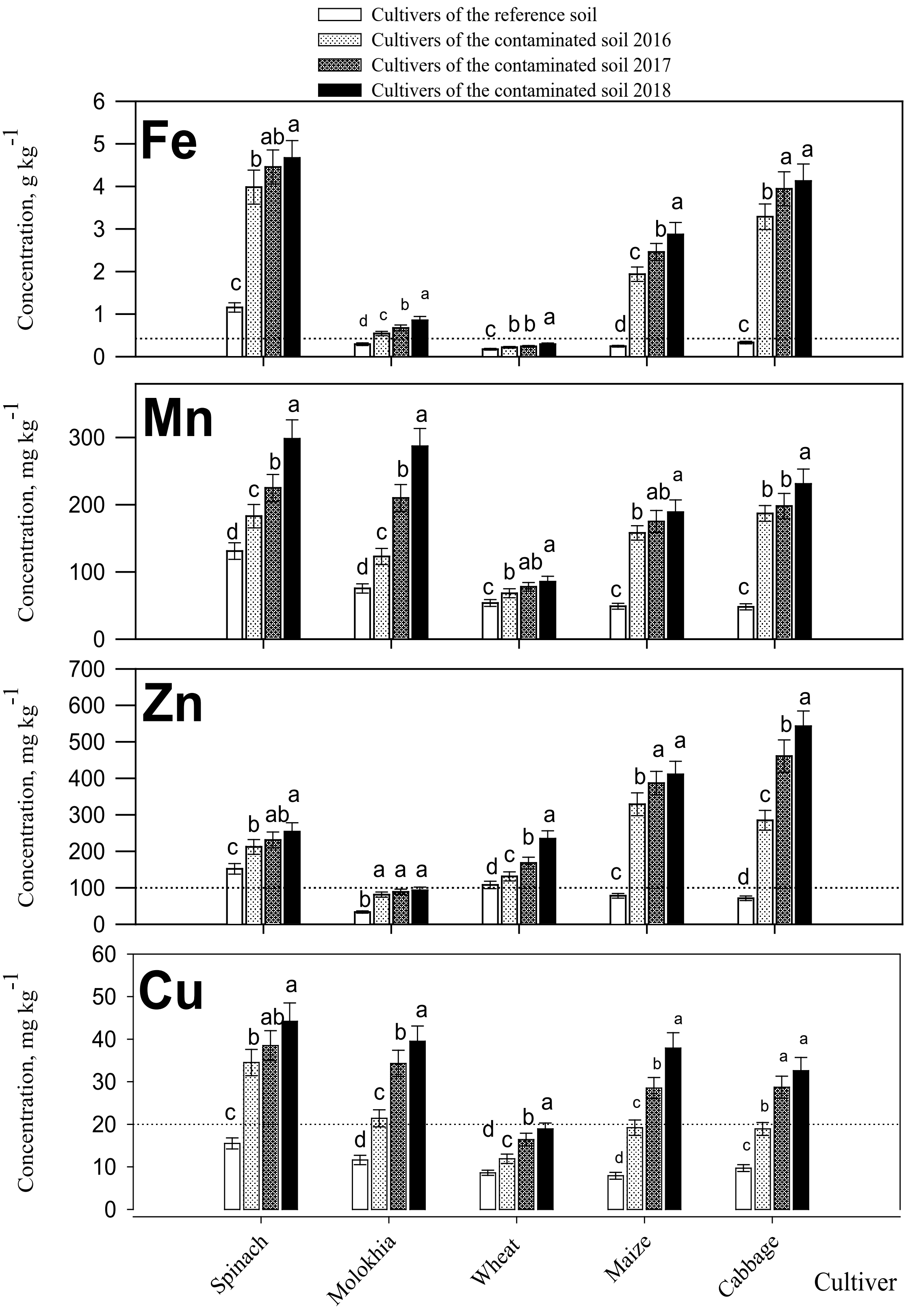

Fig. 8. Concentrations of Fe, Mn, Zn and Cu in plant shoots of the contaminated area nearby the drain (maximum acceptable level (MAC) was shown by the dot-line)

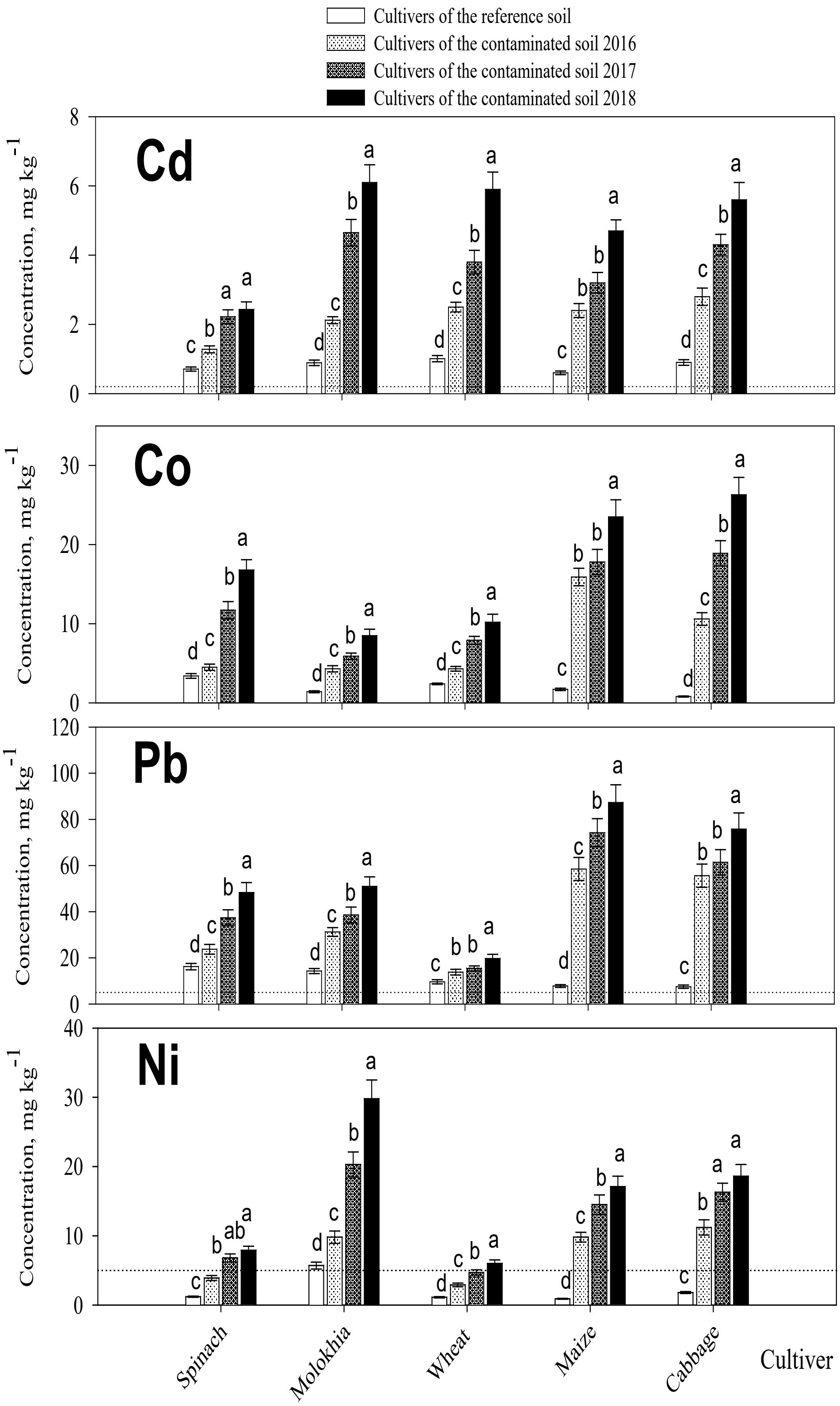

Fig. 9. Concentrations of Cd, Co, Pb and Ni in plant shoots of the contaminated area nearby the drain (dot-lines symbolize the normal ranges of PTEs in plants)

Levels of soil pollution with PTEs in relation with increasing the distance from the main drain

Levels of soil pollution with PTEs were calculated with increasing the distance from the source of pollution. Probably, hydraulic continuity decreases with increasing the distance between the main drain and the nearby arable lands. To test this assumption, concentrations of PTEs were determined within the different soil depths at four different locations (250 m apart from each other) on the other side of the main drain. Results obtained herein reveal that the levels of soil pollution with PTEs decreased considerably (within the different soil depths) with increasing the distance of land from the main drain. Soil pollution index was then calculated as the ratio between total concentrations of PTEs, within the different soil layers, divided by their background levels. The obtained values were represented graphically in Fig 10 and compared with the intervals of soil pollution index suggested by Lactusu (2000). Results reveal that soils nearby the main drain suffer from moderate pollution with Mn and Ni; whereas, the corresponding pollution levels seemed to be slight at 1000 m distance away from the wastewater drain. The nearby lands (0-30 cm surface layer) also suffer from severe pollution with Zn, Cu, Cd, Co and Pb. These concentrations represent moderate pollution levels within the surface 0-30 cm layer at 1000 m apart from the main drain. In case of Fe, all soil locations were slightly polluted with Fe. Therefore, we accept the fifth hypothesis, H5.

Conclusion and Recommendations

Concentrations of Mn, Cd, Co and Ni in shallow wells exceeded the permissible levels presented by the ECP 501 (2015) for using treated wastewater in irrigation. This water seemed to be moderately polluted with PTEs during 2016 and 2017; however, became severely polluted with PTEs during 2018. It is worthy to mention that there were no significant variations detected in CPI values between waters of the shallow wells and the corresponding ones of nearby drain water. In case of BOD, COD and OPI, their calculated values in either the irrigation well water or the drain water were higher than their acceptable levels with no significant variations in BOD between these two sources of water. This indicates that the collected water samples were severely contaminated with organic pollutants. Also, indirect hydraulic continuity probably existed between the wastewater in the main drain (the non-point source of soil pollution) and the underground (well) water of the nearby arable lands.

Using such well water for irrigation resulted in concurrent increases in both AB-DTPA and total contents of PTEs within the different soil layers during the three successive years of study i.e. 2016, 2017 and 2018. The highest concentrations of these metals were detected within the top (0-15 cm) soil layer while the corresponding concentrations seemed to be relatively lower within the subsequent soil layers. Plants grown on the nearby polluted area accumulated high concentrations of PTEs in their shoots especially Cd and Pb whose contents exceeded the permissible levels in edible leafy vegetables. On the other hand, the levels of soil pollution with PTEs decreased significantly with increasing the distance of soil from the source of pollution (drain water) while recorded slight to moderate levels of soil pollution at 1000 m distance apart from the wastewater drain. Thus, the discharge of the untreated wastewaters might possess high ecological risks even if these waters are not used for crop irrigation. It can; therefore, be deduced that the activity of industrialization brings up death to the surroundings living organisms unless certain precautions summarized in the following should be followed:

1- The industrial area of Shobra El-Khema should be transferred to new desert lands away from the arable lands.

2- Country codes should consider the quality of factory discharges within the industrial areas because of their negative implication on the surrounding environment.

3- More studies are needed to quantify the indirect implications of industrial discharges on the environment. In this concern, factories should pay for conducting such environmental studies.

4- Factories should donate or pay periodically to support the health services within the industrial area (environmental safety).

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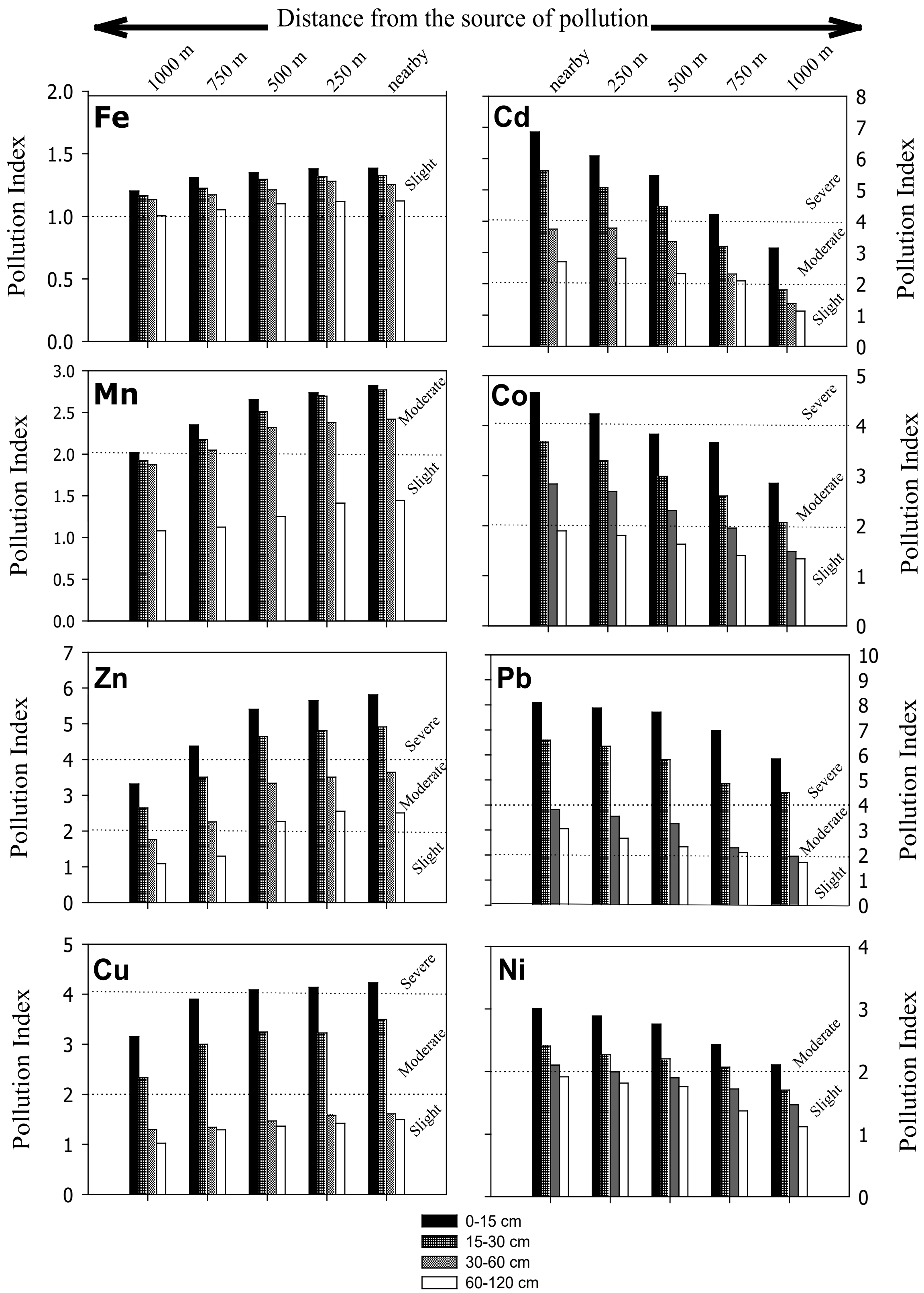

Fig. 10. Soil pollution index as affected by the distance from the main drain (dot-lines indicate the intervals of the pollution indexes)

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المخاطر البيئية المصاحبة لتسريبات مياه الصرف غير المعالجة في المناطق الصناعية

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اتجهت العديد من الدول حول العالم إلي اعادة استخدام مياه الصرف المعالج في ري المزروعات، وذلك للتغلب علي مشكلة نقص المياه بها، وفي مقابل ذلك أهتمت الحكومة بالتشريعات لمراقبة نوعية مياه الصرف المعالج بها واستخدماته في انتاج الغذاء، ورغم ذلك لم تتمكن تلك التشريعات من التعامل بصورة كفء مع التداعيات السلبية لهذه المياه علي البيئة المحيطة، ومن هنا تأتي اهمية الدراسة التالية والتي تسلط الضوءعلى منطقة شبرا الخيمة (القاهرة)، كأحد أهم المناطق الصناعية في القاهرة والتي تستقبل قنوات صرف الاراضي الزراعية المجاورة لها كميات كبيرة من مخلفات تلك المصانع غير المعالجة، وعلي الرغم من قيام الدولة بحظر استخدام مياه المصارف في ري الاراضي بهذه المنطقة، إلا ان هذه المياه لاتزال تمثل تهديد كبير للبيئة المحيطة إما بصورة سريعة أو على المدي البعيد ، نظرا لقيام العديد من المزارعين باستخدام مياه الابار في ري اراضيهم والتي تعتبرآبار سطحية، ومن المحتمل انها تستقبل التسريبات الناتجة عن تلك المصارف، ولدراسة الوضع البيئي بهذه المنطقة، فإنه تم جمع عينات المياه، والتربة والنبات من المناطق المزروعة بجوار المصارف، واجريت التحليلات اللازمة لمعرفة محتواها من معادن الحديد، المنجنيز،الزنك، النحاس، الكادميوم، الكوبلت، النيكل والرصاص (الكلية والميسر)، ومن خلال النتائج المتحصل عليها، تم حساب معامل التلوث الاجمالي للمياه (CPI) للعناصرمحتملة السمية خلال موسمي الشتاء والصيف علي مدار ثلاث سنوات متتالية وهي ٢٠١٦، و٢٠١٧ و٢٠١٨، واظهرت النتائج عدم وجود فروق ذات دلاله احصائية في قيم معاملات التلوث الاجمالي للمياه بين مياه الابار السطحية، ومياه المصارف ومع استخدام هذه النوعية من المياه في ري الاراضي المجاورة، وجد ارتفاع كل من المحتوي الكلي والميسر (المستخلص بواسطة AB-DTPA)عند مستوي دلالة ٠,٠٥ في الطبقة السطحية من التربة خلال سنوات الدراسة الثلاثة، بينما كان تركيز هذه الملوثات اقل في الطبقات التحت سطحية، وبتحليل النباتات النامية، اتضح ان النباتات الورقية تحتوي علي تركيز عالي من هذه الملوثات، خاصة عنصري الرصاص والكادميوم، والتي تعدي تركيزهما الحدود المسموح بهما، كما اكدت الدراسة علي ان مستويات تلوث التربة بالعناصر محتملة السمية قد انخفض بصورة ملحوظة مع زيادة بعد الارض عن مياه المصرف (مصدر التلوث الرئيسي) ومع ذلك سجلت الارضي التي تبعد ١٠٠٠ متر عن المصرف مستويات منخفضة او متوسطة من التلوث بالعناصرمحتملة السمية، وهذه النتيجة تؤكد علي وجود اتصال هيدروليكي بين المياه الجوفية (مياه الابار) مع مياه المصارف والذي يكون له تداعيات سلبية علي النظام البيئي المحيط لمسافة لا تقل عن ١٠٠٠متر بعيدا عن المصرف.