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Research Article

Kinetic Assessment of Potential Toxic Elements Desorption from Contaminated Soil Ecosystems Irrigated with Low Quality Water

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Abstract

Objective: The rates of some Potential Toxic Elements (PTEs) release from three contaminated soil ecosystems with high zinc equivalent values were kinetically compared to select the best remediative management technique(s). **Methodology:** The sampled soil ecosystems were irrigated with either industrial effluent for more than 80 years (Kafr El-Sheikh site), sewage effluent for more than 50 years (Konbera site) or drainage water mixed with river Nile water for more than 35 years (Sinai site). A kinetic study of PTEs were applied using two empirical models i.e., Modified Freundlich Elovich (MFE), Hoerl and theoretical models, i.e., diffusion and 1st order equations were applied to rate PTEs desorption. **Results:** All trailed models well described the rate of PTEs desorption from the studied soil ecosystems. According to the rate constants, industrial effluents could be the most hazard type of irrigation water on soil ecosystem, which gave highest rate values of release followed by sewage effluents and for less extent mixed water. Different mechanisms that influence the rates of PTEs desorption took place were discussed. **Conclusion:** Reuse of LQW in irrigating bears significant risks on different soil ecosystems. Remediation technique(s) should be focused particularly in alluvial clay soil ecosystems.

Key words: Low quality water, potential toxic elements desorption, kinetic models, sewage effluent, industrial effluent, drainage water

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INTRODUCTION

Due to the rigorous scarcity in water resources, Egyptian farmers were indebted to reuse low quality water contaminated with a variety of Potential Toxic Elements (PTEs) in farming. This reuse for extended periods intensified PTEs in soil ecosystems leading to serious hostile impacts outstandingly in some hot spots like Kafr El-Sheikh soil ecosystem^{1,2}, recently reclaimed soils at Sinai governorate, where the major source of irrigation is a mix of drainage and river Nile water from El-Salam canal water contaminated with a diversity of PTEs. Also, irrigation with sewage effluent at Konbera enriched the soil with wide-ranging types of organic and inorganic contaminants^{3,4}.

The mobility and fate of PTEs in studied soil ecosystems is governed to a large extent by competitive adsorption and desorption reactions at solid-water interfaces of minerals and soil organic matter⁵⁻⁷. In addition, certain slower processes like PTEs diffusion into micro porous solids and formation of PTEs bearing precipitates might lead to thoughtful fluctuations in PTEs speciation and solubility as time goes on⁸⁻¹¹. Comprehensive modeling of PTEs sorption and release in soil ecosystems is therefore, a disputing assignment that should be taken in account specifically competitive sorption, dissolution and surface precipitation, sorbent heterogeneity and kinetics¹²⁻¹⁴ and mentioned elsewhere¹¹.

Right through the preceding decades, massive studies had been done to clarify the dominant mechanisms controlling the fate and overall performance of PTEs in soil-water ecosystems. These studies focused on unfolding the sorption and transport of PTEs in laboratory, greenhouse and field experiments. However, modeling PTEs desorption in such soil and aquatic ecosystems contaminated with PTEs was scarce.

The main objectives of the present study are to kinetically compare between rates of some PTEs release from three types of contaminated soil ecosystems collected from Giza, Kafr El-Sheikh and North Sinai governorates and irrigated with non-point sources LQW, for the sake of selecting best remediative management technique(s) combating PTEs contamination.

MATERIALS AND METHODS

Water sampling: Three types of Low Quality Water (LQW) used in irrigation were collected from Kitchner drain in Kafr El-Sheikh governorate (industrial effluent), Lebini drain in Abou-Rawash village in Giza governorate (sewage effluent) and El-Salam canal in Sinai governorate (mixed low quality

water) and beside water sample collected from river Nile as a control. Different collected water were chemically analyzed and Doneen parameter was calculated by the equation $(0.5 \times \text{SO}_4^{-2} + \text{Cl}^-)$ to examine the validity of water for irrigation.

Soil sampling: Three different surface soil samples (0-30 cm) were collected, the 1st alluvial clay from Kafr El-Sheikh governorate S1 (about 134 km North of Cairo, in the Nile delta of lower Egypt, the latitude and longitude of Kafr El-Sheikh governorate is 31.3085444 and 30.8039474, respectively), the 2nd clay loam from Abou Rawash-Konbera, Giza governorate S2 (Latitude and longitude of Giza governorate is 28.7666216 and 29.2320784, respectively) and the 3rd clay loam from El-Tina plain, Sinai governorate S3 (Latitude and longitude of North Sinai governorate is 30.6084723 and 33.617577, respectively). The selection of these soil ecosystems were based on variation in their land use, biological properties and types and period of LQW used.

Desorption experiments: Kinetic experiments for the selected contaminated soil ecosystems were carried out using Electrical Stirred Flow Unit (ESFU) that gives precise information about PTEs desorption from the contaminated soil ecosystems¹⁵. Twenty grams portion of the selected soil samples were put in ESFU kinetic part and closed and 100 mL of the kinetic solution ammonium bicarbonate DTPA was added and the device switched on. According to the design of the device, the system will vigorously shake the soil and the solution samples were received after different periods ranged between 1 sec to 2880 min. Specific solution (5 mL) period were taken and the concentrations of different PTEs studied were determined.

Instrumentation and analysis of PTEs: Flame Atomic Absorption Spectrometry (FAAS) is a simple and well available technique frequently used in determining PTEs in natural aquatic samples. A Perkin-Elmer Flame Atomic Absorption Spectrometer (FAAS) and HACH DR890 colorimeter was used in this study. Atomic absorption measurements were carried out using air acetylene flame, while HACH colorimeter measurement with the provided test kits. The operating parameters for working elements were set of as recommended by the manufacturer.

Kinetic models used in fitting kinetic data: The kinetic results were matched in five models representing both empirical and theoretical patterns as follows:

$$\text{First order equation: } \text{Log} (q_i - q_e) = \text{Log} (b - a t)$$

Where:

$$\text{Elovich \acute{e}quation: } q = (1/a) \ln(ab) + (1/a) \ln t$$

$$\text{Modified Freundlich equation: } q = b t^a$$

$$\text{Parabolic diffusion equation: } q = b + a t^{1/2}$$

$$\text{Hoerl's: } q_t = a \times X^b \times E^a (a \times t)$$

where, q_j is a maximum amount of contaminants release, q_0 is a initial amount of contaminants at the time of starting, q_t is a amount of contaminants release at time t , t is a time in minute, k_1 is a rate constant of reaction in per second and b is a constants represent intensity and capacity factors.

The mathematical equations were tested by least square regression analysis for PTEs release in soil ecosystems. The conformity of release from studied soil ecosystems to a given equation was based on higher R^2 and lower SE values¹⁶.

Statistical analysis: All data were processed by Microsoft Excel. Regression of linear and other statistical analysis were conducted using the programs of Costate version 6.400, a statistical analysis software package published by CoHort software¹⁷.

RESULTS AND DISCUSSION

LQW characterizations: Table 1 indicate that the reused LQW is unacceptable for farming. The calculated Doneen parameters reached 18.08 meq L⁻¹ in El-Salam canal (Saini), 18.50 meq L⁻¹ in El-Lebeni drain (Konbera, Giza governorate) and 14.40 meq L⁻¹ in Kitchner drain (Kafr El-Sheik governorate), while it should not be exceed 5 meq L⁻¹. in normal conditions and for safe use in irrigation.

Results emphasized that the contents of most scanned PTEs in LQW were significantly higher compared to river Nile water (Table 2). The concentration of Ni for instance, in El-Lebeni drain at Konbera (Abou-Rawash) was 0.07 ppm, despite being undetectable in river Nile water. The same holds true for Mn, Fe, Cd and Pb, which reached to 1.71 ppm in Kichener drain at Konbera.

The highest salinity level represented by electrical conductivity EC was recorded in S1 soil ecosystem (2.5 dS m⁻¹) and the lowest one in El-Salam canal (1.75 dS m⁻¹). Although the salinity level in Al-Kichener drain at Konbera (Abou-Rawash) was virtually more or less the same as found in El-Salam canal, the highest value of PTEs were detected in the 1st type of LQW (Table 1 and 2), this result may due to entering of industrial effluents in this type of water. In general, all tested LQW exhibited unsafe index for reuse in farming. Water pH in samples collected from different governorates showed slight increase compared to that collected from Nile water.

Total PTEs content and zinc equivalent values in the studied soil ecosystems: The soil quality criterion index (Zn equivalent model) was arithmetically calculated in relation to the levels of PTEs toxicity in soil ecosystem. A quality criterion index over 250 U indicates a risky situation¹⁸. It seems reasonable to state that soil contamination and high calculated ZE parameter might be ascribed to use in irrigation with LQW for extended periods as well as heavy soil texture found as in Kafr El-Sheikh and Sinai soil ecosystems. Daily enrichment of PTEs in Abou-Rawash soil ecosystem reached a risky level as far the safety of harvest is considered.

Figure 1 represents the total concentration of Zn, Cu and Ni in all examined soil ecosystems. In Sinai soil ecosystem for example, the Zn equivalent parameter as shown in Table 3 reached 325, i.e., exceeding the critical level by 125%. The same parameter values increased up to 634 in Abou-Rawash soil ecosystem and decreased to about 540 in Kafr El-Sheikh soil ecosystem.

The highest Zn equivalent value was monitored in Abou Rawash sewage soil ecosystem might be due to the daily irrigation with sewage effluent. The persistent decreases recorded in pH values as shown in Table 3 are the resultant of organic acids release associated with decomposition of organic matter, which enhances the uptake of PTEs by growing plants.

Results given in Table 3, confirm significant increase in Zn concentration compared to other pollutants with range

Table 1: Some chemical properties of irrigation water used in selected farms with land use of these farms

Sites of sampling	pH	EC (dS m ⁻¹)	TDS (ppm)	SAR	(0.5 × SO ₄ ⁻² + Cl ⁻) meq L ⁻¹
Nile water (Giza governorate)	7.50	0.49	313.6	1.60	1.32
El-Lebiny drain (Giza governorate)	7.70	2.50	1600	6.35	18.50
Kitchenr drain (Kafr El-Sheikh governorate)	7.75	1.80	1154	3.03	14.40
El-Salam canal (North Sinai governorate)	7.68	1.75	1120	4.53	18.08

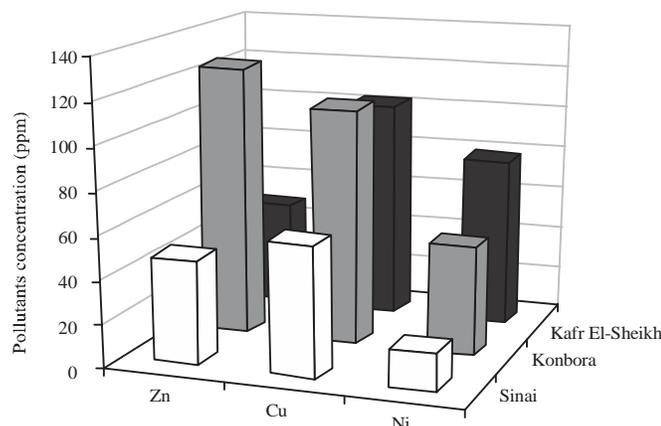


Fig. 1: Concentrations of PTE's in different studied soil ecosystems

Table 2: Heavy metals concentration in water samples collected from different sites

Sites of water sampling	Ni	Cu	Zn	Cd	Pb	Cr	Mn	Fe
Nile water (Giza governorate)	-	0.01	0.02	-	-	0.01	-	-
El-Lebiny drain (Giza governorate)	0.07	0.06	0.65	0.08	1.71	0.06	0.61	4.50
Kitchenr drain (Kafr El-Sheikh governorate)	0.25	0.98	2.22	0.04	0.02	0.08	0.52	0.56
El-Salam canal (North Sinai governorate)	0.20	0.8	3.2	0.07	5.00	0.09	0.33	5.20

Table 3: Heavy metals in soil samples collected from different sites

Sites of soil sampling	Ni	Cu	Zn	Cd	Pb	Fe	Mn	Zn equivalent
S1	22	33.8	296	18.0	43.0	460	49.8	540
S2	6	35.7	401	-	57.7	596	45.5	634
S3	18	28.0	149	17.9	25.8	150	43.0	325

S1: Kafr El-Sheikh soil ecosystem, S2: Abou-Rawash (konbera) soil ecosystem and S3: Sinai soil ecosystem

equal to 150-400 ppm, followed by Cu pollutant (26-36 ppm) with highest value calculated in Konbera soil and lowest in Sinai soil ecosystems in both pollutants. Meanwhile, Ni content in the studied soils which exhibited the lowest concentration values (6-22 ppm), the highest value determined in Kafr El-Sheikh soil that irrigated with industrial effluents and the lowest value in Konbera soil, which irrigated with sewage effluents.

The most hurt contaminants in soil ecosystems are represented by Zn, Cu and Ni, which synergistic each other. This however, diminishes soil bio-vitality that depresses microbial population compared to the individual presence of these pollutants¹.

Release of PTEs from Sinai soil ecosystem in relation to contamination level and soil characters

Conformity of used models to kinetic data: Results showed that Ni was not detected in Sinai soil ecosystem. Table 4 shows that the power function (modified Freundlich), Hoerl and Elovich kinetic equations gave high conformity in describing the kinetics of PTEs release from the studied soil ecosystems, as all showed high significance (R^2) ranged between 0.86** and 0.98**, as given in Table 4.

Elovich equation gave the highest coefficient of determination, while modified Freundlich equation gave the lowest SE value, which confirms the power function equation was the best in describing Zn, Cu and Ni desorption¹⁹. The applied models might be arranged as: Power function or MFE, Elovich and Horel's models. The fitting of more than one model in describing the kinetic data means that more than one mechanism controlled PTEs desorption from soil ecosystem¹⁶. It should be mention that 1st order model, however, gave less conformity in describing desorption data. The kinetic models used in this study are widely used in describing kinetics of PTE's release for lead²⁰, cobalt²¹ and fluorine²².

Rate constants of PTEs desorption from the studied soil samples as affected by low quality waters applied:

The rate constant a of MFE, which was found convenient to describe the rate of PTEs release showed a significant gab between the studied PTEs desorption. In case of Zn released from Sinai soil ecosystem, a constant value was 0.34 mg kg⁻¹ soil min⁻¹, while the corresponding value in Kafr El-Sheikh soil ecosystem did not exceed 0.11 mg kg⁻¹ soil min⁻¹. On the other hand, the

Table 4: Rate constants of used models describe pollutants release from different soils collected from different governorates

Governorate	Zn				Cu				Ni			
	a	b	R ²	SE	a	b	R ²	SE	a	b	R ²	SE
Modified Freundlich equation												
S1	0.11	2.41	0.97**	0.03	0.30	0.85	0.98**	0.05	0.32	0.48	0.98**	0.05
S2	1.32	2.41	0.96**	0.03	0.24	0.75	0.94**	0.05	0.33	0.49	0.95**	0.05
S3	0.34	0.98	0.94**	0.03	0.23	0.31	0.96**	0.05	0.12	0.26	0.98**	0.05
Elovich equation												
S1	37.21	29.54	0.97**	1.40	4.93	5.20	0.97**	1.52	2.78	1.63	0.95**	1.60
S2	16.53	21.5	0.96**	1.63	1.92	0.89	0.98**	1.09	2.68	1.43	0.93**	1.58
S3	0.11	4.29	0.98**	1.60	0.06	0.52	0.98**	1.56	0.85	1.61	0.95**	1.58
Parabolic Diffusion equation												
S1	11.7	275.9	0.89**	12.71	1.60	8.30	0.96**	3.01	0.93	3.03	0.98**	4.63
S2	5.73	234.05	0.86**	12.71	0.67	2.11	0.95**	3.01	0.73	2.96	0.96**	4.63
S3	1.06	9.89	0.87**	12.71	0.40	0.66	0.94**	3.01	0.72	2.47	0.98**	4.63
1st Order equation												
S1	1.59	5.72	0.64*	0.16	4.20	2.50	0.64*	0.43	4.91	1.64	0.87**	0.42
S2	0.94	5.52	0.99**	0.16	3.19	1.45	0.94*	0.43	3.81	1.74	0.97**	0.42
S3	0.88	0.92	0.66*	0.16	0.25	0.36	0.64*	0.43	1.09	4.25	0.87**	0.42
Hoerl equation												
S1	0.32	5.52	0.94**	0.03	0.40	1.90	0.97**	0.08	0.33	1.09	0.98**	0.07
S2	0.12	5.52	0.94**	0.03	0.35	1.77	0.96**	0.08	0.38	0.67	0.98**	0.07
S3	1.65	2.21	0.94**	0.03	0.42	1.08	0.97**	0.08	0.24	0.79	0.98**	0.07

S1: Kafr El-Sheikh soil ecosystem, S2: Abou-Rawash (konbera) soil ecosystem and S3: Sinai soil ecosystem

highest released Zn was found in Konbera soil ecosystem reaching 1.32 mg kg⁻¹ soil min⁻¹. The capacity factor described by b constant for Zn showed equity between Kafr El-Sheikh and Konbera soil ecosystems equal to 2.41 mg kg⁻¹, while it decreased to 0.98 mg kg⁻¹ in Sinai soil ecosystem.

Results of Elovich equation given in the same table exhibited an increase in a constant value in S1 reached 37.21 mg kg⁻¹ soil min⁻¹, a decrease to 16.53 mg kg⁻¹ soil min⁻¹ in S2 and 0.11 mg kg⁻¹ soil min⁻¹ in Sinai soil ecosystem (S3).

The diffusion mechanism also controlled the release of Zn from Sinai soil with significant R². Results given in the same table showed that the rate of release values in the model took the same trend as of Elovich. The corresponding values of intensity factor for different soil ecosystems were 11.7, 5.73 and 1.06 mg kg⁻¹ soil min⁻¹ for S1, S2 and S3, respectively. Worth to mention that Hoerl model gave a similar trend like MFE and Elovich. In general, having different models suitable to described PTEs desorption phenomena, means that these different mechanisms took place in sorption on SS, subsequently different mechanism would take place in desorption of these contaminants.

The capacity factor which represented by b constant in all used models, showed a similar ascending order order, i.e., S1>S2>S3. For example, in MFE, the best fitted model, the numerical value was 2.41 mg kg⁻¹ represented in both S1 and S2 and decreased to 0.98 mg kg⁻¹ in S3.

The rate constant of Cu desorption described by MFE showed a gradual decreasing in rate value from 0.30 in S1 to

0.23 mg kg⁻¹soil min⁻¹ in S3, with medium value equal to 0.24 mg kg⁻¹soil min⁻¹ in S2. The same trend was also showed in other models used except Hoerl model.

The capacity factor of MFE and parabolic models displayed an increasing order in S1 and decreased in S2 and lowest value was found in S3. The numerical values of capacity constant of these soil ecosystems were 0.85, 0.75 and 0.31 mg kg⁻¹, respectively. In Elovich, however, the capacity factor of S1 was 5.20 mg kg⁻¹, decreased to 0.89 mg kg⁻¹ and then increase in S3 to reach 0.52 mg kg⁻¹.

Copper desorption described by diffusion model showed an increasing rate of desorption in S1 (soil irrigated with industrial effluents), compared to other soil ecosystems reaching 1.60 mg kg⁻¹ soil min⁻¹. The capacity factor represents by b constant was also higher than in other studied soil ecosystems approaching 8.3 mg kg⁻¹ soil.

Nickel desorption from the studied soil ecosystems, which well described by the three models MFE, Elovich and Diffusion models signify that different mechanisms controlling desorption of PTEs. In MFE, the rate of Ni release from S2 was higher than either S1 or S3 with numerical values of 0.33, 0.32 and 0.12 mg kg⁻¹ soil min⁻¹ respectively. It should be mention that the capacity factor also exhibited the same trend. In Elovich and Diffusion models, the release of Ni was higher in S1 than in either S2 or S3.

In this study, the rate of PTEs desorption from the soil ecosystems governs the best remediation technique(s) that might be applied. The fate of desorption of selected PTE's is mainly depend on the mechanisms of PTEs retained in soil ecosystem directly from low quality water used in irrigation.

Table 5: Chemical properties of used soils collected from different governorates irrigated with low quality water

Soil No.	Landscape	pH	EC (dS m ⁻¹)	OM (%)	Clay (%)	Silt (%)	Texture
S1	Common bean	8.35	2.8	1.63	54.6	34.1	Clay
S2	Tomato	7.32	0.61	3.59	38	37	Clay loam
S3	Sugar beet	7.79	2.9	0.59	23.7	32	Loamy

S1: Collected from Kafr El-Sheikh governorate (Khasha village), S2: Collected from Giza governorate (Konbera village) and S3: Collected from North Sinai governorate (El-Kantara West village)

Generally, these mechanisms depend on many factors some of which related to the studied PTEs, such as their concentration, ionic strength, ionic radius, electronegative, valance charge and surface type and other characters related to the sorbents or soil system characters such as OM, pH, EC, amorphous materials and concentration of clay minerals in soil. Clay content plays an important role in accumulation of PTEs through sorption/desorption.

The PTEs inputs in the soil ecosystem with residence time, led to be retained with different mechanisms through sorption phenomena such as: (1) Adsorption which is defined as a two-dimensional uptake process governed by physical and chemical interactions between the metal and the clay surface. On clay minerals, such as montmorillonite adsorption could occur both at the edge sites, which leads to inner-sphere metal complexes and at the planar (internal) sites of the clay mineral, which results in outer-sphere metal complexes. (2) Cation exchange that involves the sorption of ions in solution onto oppositely charged, discrete sites on the surface of a soil particle. It is driven by the attractive force of maintaining electrostatic neutrality. (3) Specific adsorption that involves the exchange of PTEs and most anions with surface ligands to form partially covalent bonds with lattice ions. It results in metal ions being adsorbed to a far greater extent than would be expected from the CEC of a soil. (4) Co-precipitation that is defined as the simultaneous precipitation of a chemical agent in conjunction with other elements by any mechanism and at any rate. (5) Insoluble precipitates of PTEs metals in soils, i.e., when the physico-chemical conditions and concentrations of appropriate ions are sufficiently high many metals form insoluble precipitates, which could play a role in controlling their solubility in the soil solution. In this study, different mechanisms expected to be take place between pollutants and different ecosystems studied²³.

At heavy textured soil ecosystems, electro-negativity and ionic radius of pollutants controlled the mechanism of sorption and desorption regardless the high concentration monitored in S1. Increasing clay content in S1 compared to other soil ecosystems associated with decreasing ionic radius of Zn (1.65) might enhance Zn retention in S1 and increased in S2 as shown in rate constants of MFE. In the study²⁴, suggested that adsorption to oxide surfaces, which have a

high affinity for Zn, clays, organic matter or any combination of these might be important in controlling Zn solubility, as there are excess of oxides alluvial soils like S1. Iron, Mn and Al oxides as well have negatively charged surfaces and thus might powerfully bind metals in neutral to alkaline soils. Adsorption by these oxides had been suggested by several studies like²⁵.

On the other hand, if Zn²⁺ is adsorbed to organic matter, two other mechanisms might occur. Zinc could be adsorbed to organic matter and replace only one proton by adsorption to a carboxyl group or have an ion-exchange reaction with a zero slope such as: Zn²⁺+Ca-EXCH ~ Ca²⁺+Zn-EXCH this may explain the decreasing the rate of release from S1.

Decreasing clay content in S2 compared to S1 (Table 5), beside the daily input of pollutants in Konbera soil and enhancement Ni, Cu and Zn to each other all enhanced the release of both Ni and Zn from S2, this might be declared through having the same trend of Ni release from the same soil.

The high concentrations of readily available forms of PTEs daily thrown into the drains and reach the S1 and S2 may explain the high recorded *a* constants of Elovich equation in both soils. The *a* constant value of Elovich equation is convenient to describe the rate ions uptake by growing plants. The results documented¹⁶, found a close relationship between Fe uptake by plants and a constant of Elovich model. In addition²⁶, a significant correlation between water soluble and exchangeable forms in PTEs distribution with the same constant.

Nickel adsorption is most likely the mechanism responsible for the initial fast stage of Ni sorption and surface precipitation appears to be an important mechanism controlling the slower stage of Ni uptake, beside its low concentration compared to others as shown in Table 4 might be satisfactory reason explain the decreasing rate of release compared to other PTEs. The majority of Ni adsorption studies on variable charge system, principally to Fe and Al oxides and silicate minerals.

Horel model is convenient to describe the disability of PTEs to be desorbed to the soil ecosystem¹⁸. In different PTEs like Zn, results showed that the lowest value was found in the soil ecosystem irrigated with industrial effluents (S1), which

means that such ecosystems have high ability to desorb PTEs. Unlike S1, results indicated that the highest value was found in S3 (1.65), which means that desorption of PTEs from El-Tina plain soil is not that easy. Worth to mention that this observation was not detected in Ni desorption from any ecosystems studied. This result may due to Ni have highest electro negativity 1.91 Pauling and biggest atomic radius compared to other pollutants.

In this study²⁷, showed that Ni specific adsorption seemed to be low and the formation of external sphere complexes was predominant mechanism of Ni adsorption.

CONCLUSION

Reuse of LQW in irrigating bears significant risks on different soil ecosystems. From kinetic prospective PTEs desorption from different soil ecosystems trailed in this study, industrial effluents is the most dangerous compared to sewage effluents or mixed drainage water with river Nile water. Remediation technique(s) should be focused on alluvial clay soil ecosystems receiving LQW for extended periods since it gave significant high rates of PTEs desorption compared to other soil ecosystems tested.

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