# Minimizing Hazards of Heavy Metals in Vegetable Farms using Phytoremediation Technique

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## ABSTRACT

**Two techniques were applied in this work to minimize the hazards of potentially toxic elements (PTEs) in three contaminated soils namely; chemical remediation represented by rock phosphate (PR) with phosphate dissolving bacteria (PDB), Montmorillonite modified clay mineral (MCM) and mixture of both chemical remediated materials, and intercropping technique represented by Radish (*Raphanus raphanistrum*) and Turnip (*Brassica rapa*). The selected soils suffered from different sources of pollution and varied types of heavy metals emitted to the agricultural ecosystem. Tomato (*Solanum lycopersicum*) plants as an important economic agricultural crop in the studied region were taken in this work as an indicator to evaluate the applied techniques to remediate heavy metals pollution in soils textured from sandy clay loam to clay. The obtained results imply all techniques significantly decreased the concentration of Pb, Cd, Ni, Zn and Cu in the soils, however, chemical remediation technique especially the mixture between the two treatments (T3) were more effective in decreasing the hazards of pollutants in the studied soils. Different mechanisms between both used techniques and heavy metals were discussed.**

**Keywords: Phytoextraction; Heavy metals; Intercropping; Tomato; Radish; Turnip.**

## INTRODUCTION

Soil contamination from heavy metals is one of the most serious environmental problems in Egypt. There are many sources of heavy metals emissions in agricultural soils in Egypt such as industrial activities, mining and smelting of metalliferous ores, electroplating, gas exhausts, energy and fuel production, fertilizer and pesticide applications as well as municipal waste, could be more serious in soil ecosystems. Metal pollutants can easily enter the food chain if heavy metal-contaminated soils were used for the production of food crops and create healthy problems (Wang et al. 2003). Investigating the fate and effects of metals in components of ecosystems are great significance (Swaileh *et al.* 2001). Numerous tests were carried out including leaching, replacing contaminated soil, applying soil amendments (Wang et al. 2003) and remediation (Ximenez-Embum *et al.* 2001). However, these are expensive procedures and produced irreversible damage to the environment (Ximenez Embum *et al.* 2001). The intercropping technique is a system in which two or more different crops (annual or perennial) are grown simultaneously on the same plot of land. This practice is traditional in the tropics and is starting to be used in temperate climates for environmental purposes. Phytoremediation is a biology-based technology considered as a novel approach and one of the management practices for the bioremediation of contaminated sites and the intercropping technique could be used in remediation. Phytoremediation is friendly and potentially cost-effectiveness, and affordable biological and technological solution with long-term application (Ximenez–Embum *et al.* 2001). Laghlimi et al. 2015. Mahmoud and Hamza, 2017 defined phytoremediation as the process, which uses green plants for the relief, transfer, stabilization or degradation of contaminants from soil, sediments, surface waters, and groundwater. Radish (Raphanus sativus) and Turnip (Brassica rapa) are annual vegetables found throughout the world and is now commonly recognized as small bulbous red root. Although Brassica rapa (Turnip) is considered as one of the most important Brassica species consumed all over the world and is an important part of the human diet, very few studies revealed the PTEs contents in Brassica rapa, (Wang et al., 2015). Mourato et al. (2015) confirmed that Brassica sp. hyperaccumulated up to 40,000 mg kg-1 Zn and up to 2000 mg kg-1 Cd in their shoot dry matter without showing any toxicity symptoms on the plant, in conclusion, these plants characterized with: (i) their ability to accumulate PTEs in the aboveground parts; (ii) tolerance to the high PTEs concentrations in soil; (iii) fast growth and high accumulating biomass; (iv) easy growing and harvestable. Chemical stabilization involves fixing up the contaminants in stable sites by mixing or injecting organic or inorganic soil amendments such as clay minerals, phosphates, lime, organic materials, iron and manganese oxides and coal fly ashes., etc Bolan et al. (2003). The in situ immobilization of PTEs in soils, using amendments, such as apatite, clay minerals or

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waste by-products were considered as a promising alternative to the currently available remediation methods. The main advantage of the stabilization approach is the simple mixing of amendments with soil, using common agricultural facilities, or placing it as a liner around the contaminated location (Usman et al., 2006) Several soil additions had been tested by (Fawzy and Rashed, 2012) such as rock phosphate, bentonite and kaolinite to reduce PTEs availability in soils. However, from an economic evaluation point of view, such treatment/removal technologies were generally costly and destructive (Koptsik, 2014). Recently, Bolan et al. (2014) discovered that immobilizing amendments such of precipitating agents and sorbent materials decreased the bio-availability and mobility of metals or metalloids from soil solution either through adsorption, complexion or precipitation reactions. Immobilizing agents could be used to reduce the transfer of metals or metalloids to food chain via plant uptake. However, the long-term stability of the immobilized metals or metalloids should be monitored. Essa and Farragallah (2006) stated that clay minerals are required to inhibit the migration of contaminants from the soil ecosystem to the surrounding environment. They were among the major materials that interact with almost all soil contaminants. Clays often represent a short-term sink of PTEs in soils, because of their adsorptive properties. Asaad et al. (2013) mentioned that natural and modified clay minerals had been studied as adsorbents for the removal of various toxic and hazardous contaminants of major concern to the environment. Montmorillonite showed good absorbance for removal of toxic PTEs such as Cd, Cr, Co, Cu, Fe, Pb, Mn, Ni and Zn (Satje and Nelson, 2009). In general, both techniques have advantage and disadvantages in their application in contaminated soils. Although there is voluminous research about minimizing the hazards of PTEs by different technique(s), a little was taken the comparison between these techniques to choose the best in minimizing the hazards of environmental pollution. The aim of the present work was to compare between chemical remediation represented by a single application of rook phosphate and Montmorillonite or a mixture of both and intercropping technique represented by Radish and Turnip cultivated with tomato as a test plant on minimizing the hazards of PTE's in alluvial soils.

## MATERIALS AND METHODS

### Soil Sample:

### Area of study:

Sahl El Husseiniya region, El-Sharqia Governorate is irrigating with water of Bahr El-Baqar drain. The selected cultivated land is located between latitudes N 30° 50' 59.8" and longitudes E 31° 59' 05". Some physic-chemical properties and total heavy metals contents of the studied soils are present in (Tables 1 and 2).

### Samples preparation:

Soil samples (0-30 cm) were air dried, crushed gently and sieved through a 2 mm sieve prior to analysis. The main soil chemical and physical properties were determined as described in Page et al. (1982) and the obtained results presented in Table (1). A part of soil samples (1 g) were digested according to Shumo et al. (2014) using $HNO_3$ and $H_2O_2$ mixture for analyses of the total content of heavy metals. After harvesting, plant samples were oven-dried at 55°C and ground to fine particles; it should be mention that all treatments had three replicates. After harvest, Radish and Turnip plants were dried at 70°C, weighed and wet-ashes using a 5:1 $HNO_3$: $HC1O_4$ acid mixture. The concentrations of the heavy metals in the clear-acid extracts were determined. Heavy metals concentration in soil and plant samples was measured using Inductively Coupled Argon Plasma, (ICAP 6500 Duo, Thermo Scientific, England).

**Table 1. Some physical and chemical properties of the studied soil samples**

| Particle size distribution (%) | | | | OM | $CaCO_3$ | CEC | EC | | Soil |
|---|---|---|---|---|---|---|---|---|---|
| Texture | Clay | Silt | Sand | (%) | (%) | (cmol $Kg^{-1}$) | ($dSm^{-1}$) | pH | No |
| Sandy clay loam | 23.20 | 18.00 | 58.80 | 2.81 | 4.60 | 29.70 | 0.72 | 7.01 | S1 |
| Clay | 45.80 | 14.00 | 40.80 | 3.19 | 5.06 | 41.18 | 2.29 | 7.35 | S2 |
| Clay | 61.58 | 27.76 | 10.48 | 7.97 | 3.27 | 96.00 | 6.64 | 7.38 | S3 |

**Table 2. Total concentrations (μg/g) of potential toxic elements in the studied soils**

| Soil No. | Pb | Cd | Ni | Cu | Zn |
|---|---|---|---|---|---|
| S1 | 42.44 | 10.67 | 24.81 | 32.91 | 72.02 |
| S2 | 46.04 | 12.82 | 62.16 | 44.94 | 84.87 |
| S3 | 57.08 | 15.9 | 79.45 | 108.96 | 162.74 |
| FAO/WHO (2001) | 10 | 3 | 50 | 100 | 60 |

1000 mg/l multi-element certified standard solution (Merck, Germany) was used as stock solution for instrument standardization.

### Greenhouse experiment steps

### Intercropping Plant system used in soil remediation using Tomato and hyper accumulator

#### Experimental Design

In a completely randomized experimental design at the greenhouse of the Desert Research Centre (DRC), Tomato (Solanum lycopersicum), and two hyper accumulator plants namely; Radish (*Raphanus sativus*) and Turnip (*Brassica rapa)* were planted and grown in three replicates to investigate their efficiency in minimizing the adverse impacts of prolonged irrigation with low quality water on the selected test soils. For each types of soils, sufficent numbers of plastic pots were filled with 5 kg soil/pot for Tomato and Radish seeds system (TR) plants growth and with 7 kg soil/pot for Turnip and tomato seeds system (TT).

#### Applied chemical remediation materials:

Two types of control were used for each collected soils represented by:

1.(C): Pots filled with untreated with remediation and cultivated soils with Turnip or Radish as used hyper-accumulators plants in this work with Tomato.

2.(T1): Soil treated with Rock phosphate (PR) mixed with Phosphate Dissolving Bacteria (PDB) at the rate of 1.25 ton fed$^{-1}$ and cultivated with seeds of tomato (*Solanum lycopersicum*) and Radish (*Raphanus sativus*) or Turnip (*Brassica rapa)*.

3. (T2): Soil treated with modified Montmorillonite clay mineral (MCM) at the rate of 1.25 ton fed$^{-1}$ with Tomato and Radish (TR) or Tomato and Turnip (TT).

4. (T3): Soil treated with mixture of both chemical remediated materials at the rate of 1.25 ton fed$^{-1}$ with Tomato- Radish (TR) or Tomato –Turnip (TT).

Through the growing season (160 day) and accrding to variations of soil properies and water requirements, the plots were irrigated at 60% of their water holding capacities with Bahr El-Bakar low quality water. Daily observations were conducted and the apparent differences of the plants were noted. At maturity stage, hyperaccumulator plant samples were gentely separated from soil, analyzed to their vegetative characters, kept in paper bags. The plant samples then put in an oven at 70ºC for 3 days and then analyzed for their PTEs contents. All collected soil samples were air dried and prepared to different physical and chemical analysis.

#### Plant analysis

All treatments were replicated four times. After harvest, Tomato, Radish and Turnip plants in greenhouse experiments were dried at 70 °C to a constant weight, grounded to fine powder, and dry-ached in a muffle furnace at 500$^0$C for 6 hr. the ash was dissolved in a mixture of 2 M HCL and 1M $HNO_3$ (Nanda Kumar et al., 1995). The concentrations of the heavy metals in the clear-acid extracts were measured using ICP.

#### Statistical Analysis:

Statistical analysis was done using SAS software to evaluate the applied remediation amendments in minimizing the hazards of heavy metals in used contaminated soil ecosystem.

## RESULTS AND DISCUSSION

#### Heavy metals status in used soils

Results in the Table (2) imply that the range of the total concentrations of heavy metals in the studied soils ranged between 10 and 16 µg/g for Cd; 72 to 163 µg/g for Zn; 32 to 109 µg/g for Cu; 42 to 58 µg/g soil for Pb. In other words, the descending order of heavy metals concentration in soil is:

Zn > Pb> Cu> Ni > Cd

According to documents of FAO/WHO guidelines (FAO/WHO, 2001) were shown in Table (2), Pb concentrations in all used soils are higher than FAO/WHO level. In addition, data showed that S3 is higher than S1 and S2. According to Bodek et al., (1988), Pb concentration could vary significantly, even within localized areas, due to soil types and the presence of other sources. Concerning Zn concentrations in soils, the results indicated that Zn concentrations are higher than that of FAO/WHO values (60 µg/g) in all the used soils. In general Zn usually remains adsorbed to the soil. In some studies; however, at waste disposal sites, Zn could be leaching (Saber, et al., 2012). Data showed increasing of Zn concentrations than the safe limits in all used soils, this result might be due to the continuous addition of low quality waters and also may be due to the low leaching of heavy metals into the deeper layers of the soils or reached groundwater. Results in Table (2), indicated that Cu concentrations in the studied soils were less than the range values documented by FAO/WHO for S1 and S2 with exception found in S3 Value. Most of the copper compounds will be settled and bounded to water, sediments or soil particles. Also, the concentrations of copper were high in S3 due to the heavy texture of used soils and the contentious use of Bahr EL-Baqar water in irrigation for extended periods; these results directly led to accumulation of Cu. It should be mention that Cu removal by the edible crops planted in these areas is the main reason of decreasing soil Cu concentration in some light texture soils and also due to narrow leaching of heavy metals into the sub-soils layers or even to the groundwater. Cadmium concentrations in the studied soils are high in the soils and higher than that in FAO/WHO value. The high concentration was found in S3 compared to S1 and S2. This result may due to increasing of clay content and organic matter of S3. Nickel concentrations in the soil samples (Table 2) are higher in their values compared to FAO/WHO values in S2, S3 with exception found in S1. The irrigation by Bahr El-Baqar water and heavily uses of agricultural fertilizers especially phosphates could be the main source of Ni accumulation, however, it is unlikely to build-up in the soil in the long term from their use (McGRATH , 1995). Results represent in Table (2) also showed that Cd concentration is higher than that of average upper earth crust (Wedepohl , 1995). Naturally, Cd occurs in soils as a result of the weathering of the parent rock (Alloway, 1995). Although most natural soils contain less than 1 µg/g cadmium from the weathering of parent materials, those developed on black shales and associated with mineralized deposits can have much higher levels (Alloway, 1995). Atmospheric deposition, phosphatic fertilizers are important sources of cadmium pollution (Alloway, 1995 and ATSDR, 2008). Surface soils commonly contain higher concentrations of Cd than subsurface horizons. The higher concentrations of Cd in surface horizons are probably due to the cycling of Cd from lower depths to the surface by plants (Page, et al., 1987). Cadmium was higher in the study area due to the uses of phosphatic fertilizers, irrigation by water of Bahr El-Baqar Darin.

**Chemical remediation of contaminated soils**

Results in the Table (3) showed that the application of Rock phosphate PR (T1) with PDB (Phosphate Dissolving Bacteria), T1, decreased Pb concentration in S1 significantly from 42.44 to 23.1 µg/g. The corresponding values of the same treatment were about 46.04 µg/g in control decreased to 19.80; and from 57.08 in control to 16.5µg/g for control, S2 and S3 respectively. As shown in the table, although the same treatment significantly decreased the other pollutants found in used soils, some values still higher than FAO/WHO critical values. For example, the soils treated with T1 treatment decreased Cd concentration to about 6.4, 5.5 and 4.6 µg/g for S1, S2 and S3. It should be mention that according to FAO/WHO, the critical value is 3. Rock phosphate (PR) or soluble phosphate sources were widely investigated as a means of chemical treatment for the conversion of heavy metals into insoluble compounds. Phosphoric acid, ammonium, sodium and potassium phosphate react with multivalent cations to form insoluble orthophosphates. Several hydrogen or dihydrogen phosphates have been investigated also with the aim of scavenging soluble heavy metals. The final products of the reactions of soluble phosphates with heavy metal ions are insoluble compounds which are stable in geological environments (Saber, et al., 2012). The Results also showed that application of remediative material significantly decreased Pb concentration by about 62% in S1, this percentage value increased to 70 and 80% in S2 and S3 regardless the treatment s applied. Application of clay mineral (T2) showed variations in percentage of decrease for different pollutants. It is clear that T2 treatment decreased Cd in soils by the ranges between 54-84%, Ni 26-83%, Cu 18-82%, Zn 85-87% of total concentrations of pollutants, it should be mention that the highest percentages of pollutants retention were almost observed in S3. Clay, a small particle, found naturally on the surface of the earth composed mainly of silica, alumina, water and weathered rock (Kennedy, 1990). According to Okoro et al., (2012), PTEs incorporated with clay mineral seemed to be relatively inert and less available (non-labile forms). In addition, the in situ immobilization of PTEs in soils, using amendments, such as apatite, clay minerals or waste by-products were considered as a promising alternative to the currently available remediation methods, the main advantage of the stabilization approach is the simple mixing of amendments with soil, using common agricultural facilities, or placing it as a liner around the contaminated location (Usman et al., 2006). The mixture of both treatments applied to contaminated soil (T3) was the best treatment in minimizing the hazards of inorganic pollutants in the studied soils. Results in the table 3 showed that application of T3 decreased Pb from 42 µg/g in control to about 6 µg/g in S1, 5.2 µg/g in S2 and 4.5 µg/g in S3 to reach the save level according to WHO/FAO value. Although the same trend was observed for other elements in the soils, results showed that application of clay mineral mixed with PR was significantly valued treatment to Pb, Cd and Zn compared to Ni and Cu pollutants. For example, application of T3 decreased Pb, Cd and Zn by about 86, 85 and 83% in S1 compared to control (untreated soil), while the decreasing order values were about 13 and 13.2 % for both Cu and Ni. The same trend was also observed in others soils with variations in percentage of decrease mainly due to the percentage of clay contents in the used soils and also may due to the chemical properties of the studied soils.

**Table 3. Total concentrations of potential toxic elements in the studied soils after chemical remediation treatments applied**

| Remediation / Treatments | Pb | Cd | Ni | Cu | Zn |
|---|---|---|---|---|---|
| | | S1 | | | |
| Cont.1 | 42.44 | 10.67 | 24.81 | 32.91 | 72.02 |
| T1 | 23.1 | 6.44 | 14.14 | 25.9 | 42.7 |
| T2 | 16.31 | 4.9 | 18.48 | 27.08 | 30.1 |
| T3 | 6.02 | 1.55 | 21.69 | 28.56 | 12.46 |
| | | S2 | | | |
| Cont.2 | 46.04 | 12.82 | 62.16 | 44.94 | 84.87 |
| T1 | 19.80 | 5.52 | 12.12 | 22.20 | 36.60 |
| T2 | 13.98 | 4.20 | 15.84 | 23.21 | 25.80 |
| T3 | 5.22 | 1.33 | 18.59 | 24.48 | 10.68 |
| | | S3 | | | |
| Cont.3 | 57.08 | 15.9 | 79.45 | 108.96 | 162.74 |
| T1 | 16.5 | 4.6 | 10.1 | 18.5 | 30.5 |
| T2 | 11.65 | 3.5 | 13.2 | 19.34 | 21.5 |
| T3 | 4.35 | 1.11 | 15.49 | 20.4 | 8.9 |
| FAO/WHO (2001) | 10 | 3 | 50 | 100 | 60 |

Saha et al. (2002) explained that at low metal concentrations in soils, metals are mainly adsorbed onto specific adsorption sites, while at higher metal inputs soils lose some of their ability to bind heavy metals as adsorption overlap, becoming thus less specific for a particular metal. In other words, metal sorption becomes unspecific at higher metal concentrations, when the specific bonding sites become increasingly occupied, resulting in lower Kd values (Basta and Tabatabai, 1992; Yu et al., 2002; Sastre et al., 2006). Table (4) represents the effects of remediation materials applied on heavy metals concentration in tomato plants cultivated in chemical remediated soils. These are significant decrease in all pollutants studied compared to control. Application of PR fortified by phosphate dissolving bacteria (PDB) decreased Pb from 0.5 µg/g to 0.35 µg/g in S1, the corresponding values for other soils were 0.35 decreased to 0.25 and 0.27 decreased to 0.14 µg/g in S2 and S3, respectively.

**Table 4. Heavy metals content µg/g in tomato plants cultivated in chemical remediated soils**

| | Pb | Cd | Ni | Cu | Zn |
|---|---|---|---|---|---|
| | | S1 | | | |
| Control | 0.50 | 1.75 | 1.63 | 3.33 | 6.41 |
| T1 | 0.35 | 1.22 | 1.52 | 2.33 | 4.48 |
| T2 | 0.24 | 0.93 | 1.39 | 2.44 | 3.16 |
| T3 | 0.09 | 0.30 | 1.06 | 2.57 | 1.31 |
| | | S2 | | | |
| Control | 0.35 | 1.33 | 1.98 | 3.48 | 4.52 |
| T1 | 0.25 | 0.87 | 0.76 | 1.67 | 3.20 |
| T2 | 0.17 | 0.67 | 0.99 | 1.74 | 2.26 |
| T3 | 0.07 | 0.21 | 1.16 | 1.84 | 0.93 |
| | | S3 | | | |
| Control | 0.27 | 0.70 | 2.32 | 3.67 | 1.87 |
| T1 | 0.14 | 0.53 | 0.61 | 1.33 | 2.56 |
| T2 | 0.13 | 0.42 | 0.79 | 1.39 | 1.81 |
| T3 | 0.05 | 0.17 | 0.93 | 1.47 | 0.75 |

The same treatment also decreased the other pollutants found in plants, for example, application of T1 decreased Cd concentration in tomato plants from 1.75 to 1.22; 1.33 to 0.87 and from 0.70 to 0.53 µg/g in S1, S2 and S3 respectively, it should be mention that the same trend was also observed in other pollutants. Table 4 also indicated that using Montmorillonite clay mineral in used soils (T2) significantly decreased all pollutants in cultivated plants higher than applied PR in T1. Results in the same table showed that application of T2 decreased Zn concentration more than 50% in all used soils and about 25% under T1. Many scientists have studied the adsorption of heavy metals on clay minerals including zeolite, sepiolite, attapulgus, illite, montmorillonite and other natural clay minerals or modified forms, and have found that they have great potential to remove heavy metals from water (Liu and Gonzalez 1999; Liu 2007; Missana, et al., 2008; Bailey et al.1999; Gworek 1992a, 1992b). These clay minerals are low-cost materials and offer an attractive and inexpensive remediation option. They are abundant and cheap, with negatively charged layered aluminosilicates that make them good cationic adsorbents because of their relatively large surface areas (Wu et al. 2009). In recent years, researchers have used clay minerals in the remediation of heavy metal contaminated soil and achieved positive results (Haidouti 1997; Zorpas et al. 2000; Nissen, et al., 2000; Garćla-Sánchez, et al., 1999; Pivertz 2001). The mixture of both T1 and T2 that applied to contaminated soils (T3) gave the lowest values of pollutants found in tomato plants compared to the single face of each treatment. Application of T3 to contaminated soils led to a significant decrease of all pollutants found in soils and specifically Cd. Numerically, data in the table (4) showed that application of T3 to contaminated soils decreased Cd concentration from 1.75 to 0.3 µg/g, 1.33 to 0.21 µg/g, and from 0.70 to 0.17 µg/g in S1, S2 and S3 respectively. In addition, the lowest values observed in pollutants were found in S3, for example, application of T3 decreased Cu concentration to 2.57, 1.84 and 1.47 µg/g in S1, S2 and S3 respectively.

**Effect of intercropping system on heavy metals concentrations in Tomato and Hyper accumulator plants.**

Figure (1) showed that Pb concentration values in tomato cultivated in TT system were 0.25, 0.28 and 0.26 µg/g in S1, S2 and S3 respectively, the corresponding values of TR system were 1.41, 1.46 and 1.47 µg/g, in S1, S2 and S3 respectively. These results indicated that accumulation of Pb in cultivated turnip plants was more than radish, numerically, Pb concentration in Turnip cultivated with radish were 3.30, 3.38 and 3.20 µg/g in S1, S2 and S3, while the corresponding values were 2.12, 2.20 and 2.21 µg/g for S1, S2 and S3 in Radish cultivated with tomato. In all cases, Pb concentration in tomato cultivated with hyper-accumulator plants was less than pollutant concentration in cultivated tomato individually which reached to 3.54, 3.66 and 3.68 µg/g in S1, S2 and S3, respectively. Although the same trend was observed in other elements, Cu and Zn (Figures 4 and 5) gave the highest values inside the plants.

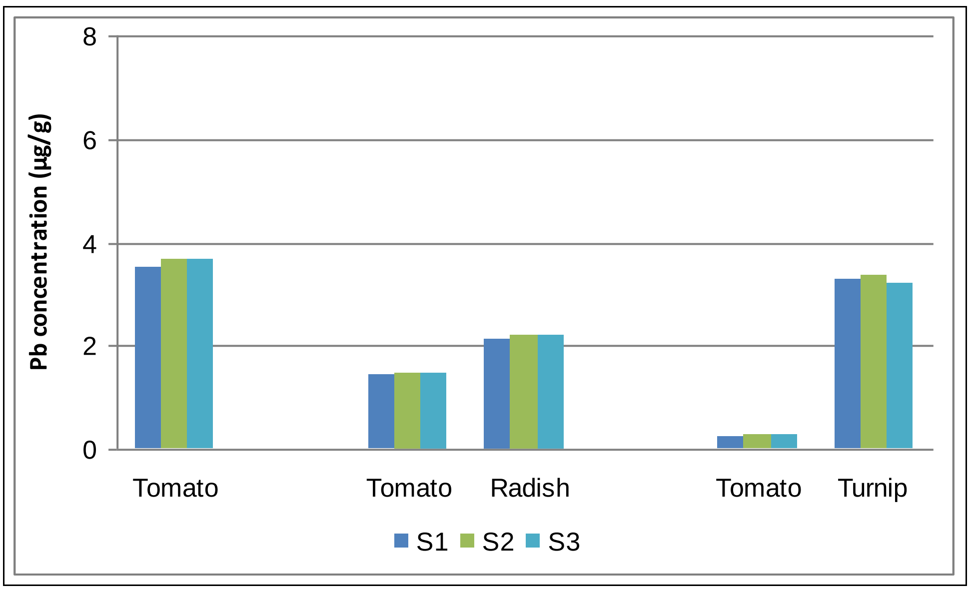


**Fig. 1. Effect of Intercropping with Radish and Turnip on Pb concentrations in Tomato plant**

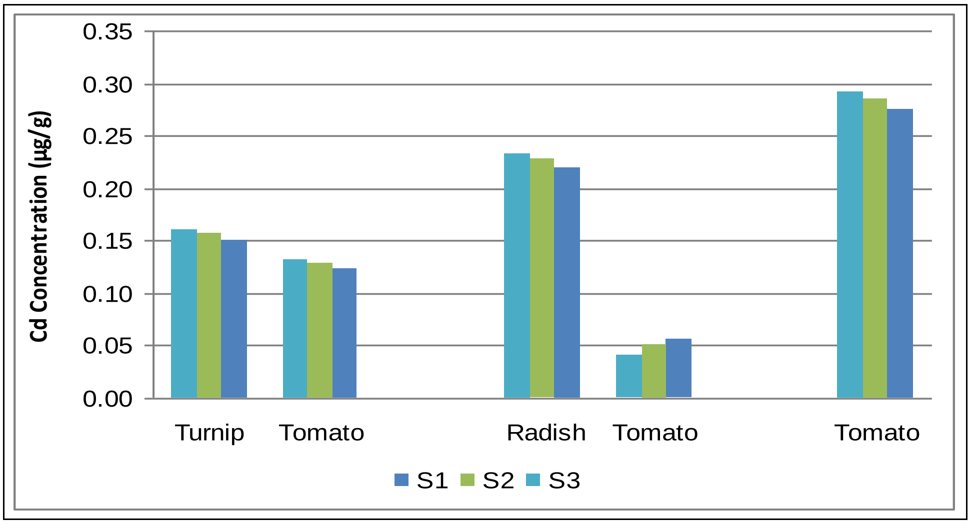


**Fig. 2. Effect of Intercropping with Radish and Turnip on Cd concentrations in Tomato plant**

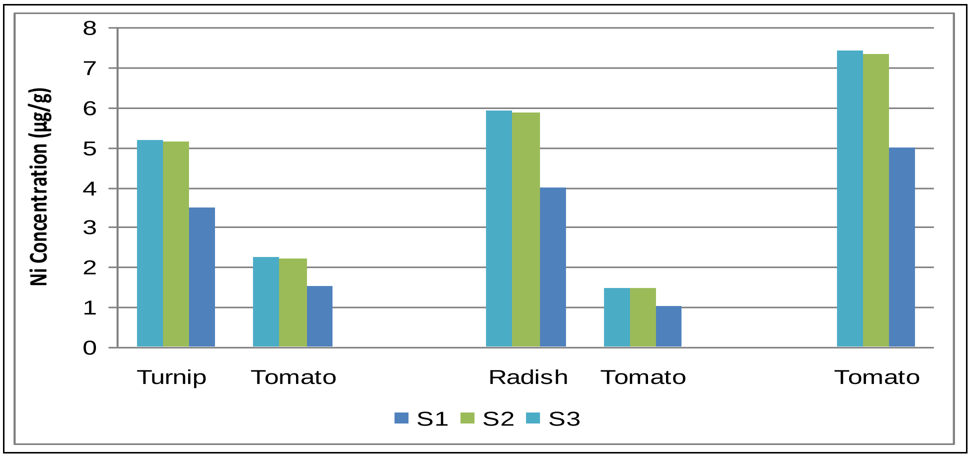


**Fig. 3. Effect of Intercropping with Radish and Turnip on Ni concentrations in Tomato plant**

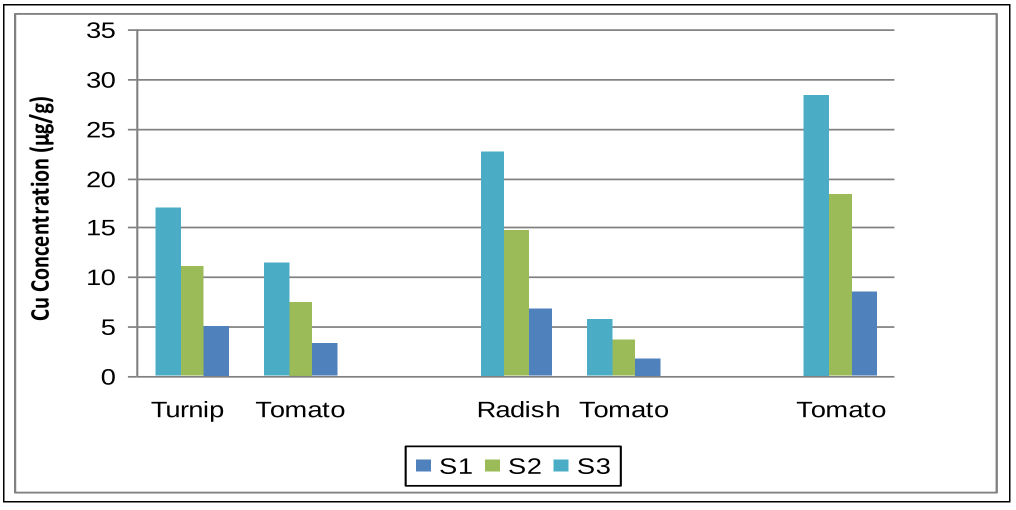


**Fig. 4. Effect of Intercropping with Radish and Turnip on Cu concentrations in Tomato plant**

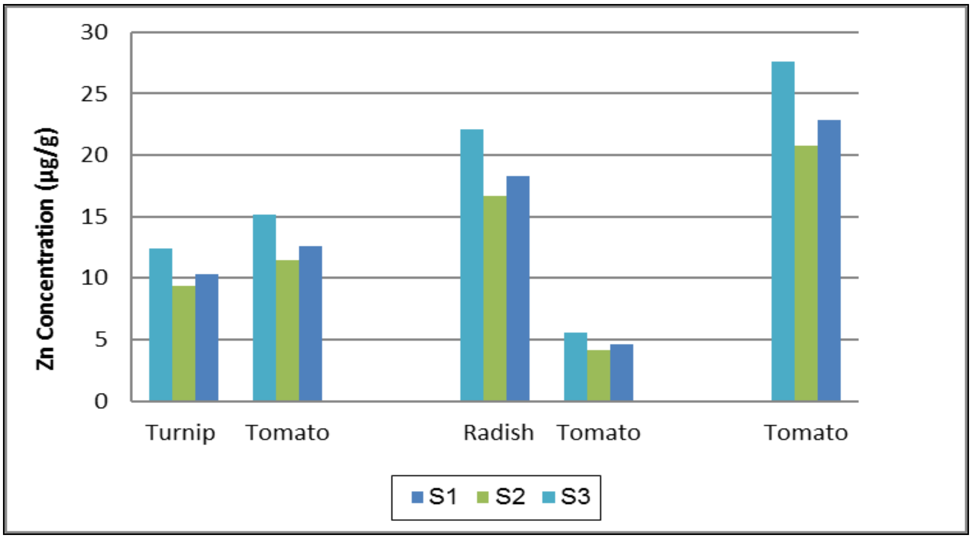


**Fig. 5. Effect of Intercropping with Radish and Turnip on Zn concentrations in Tomato plant**

The data also showed that Cu concentration in tomato plants cultivated in S1, S2 and S3 were about 8.5, 18.5 and 28 µg/g, respectively, the respective values for TR and TT were decreased to 1.7, 3.9, 5.8 and 3.8, 7.4, 11.5 µg/g in S1, S2 and S3, respectively. The highest values were observed in Zn compared to other pollutants reached 4.8, 4.2, 5.5 and 12.6, 11.5, 15.2 µg/g for TR and TT cultivated in S1, S2 and S3 respectively, it should be mention that the concentration of Zn in cultivated tomato plants individually were 23, 21 and 27.5 in S1, S2 and S3 respectively. Figures (6-10) showed that different applied intercropping systems in this work significantly clean up the used soils from the studied pollutants compared to soils cultivated with tomato only. Results showed that residual Pb concentration (figure 6) found in soils after harvesting the cultivated tomatoes individually was 42.5, 50 and 57 µg/g in S1, S2 and S3, respectively, these values decreased to about 34.5, 30 and 25.5 µg/g of S1, S2 and S3 in TR system, the corresponding values of cultivated Turnip with tomato (TT system),

**Effect of intercropping on residual elements concentrations in remediated soils**

decreased Pb concentration to about 28.5, 25 and 21 µg/g in S1, S2 and S3, respectively. This result was expected for seeing that Pb concentration inside Turnip plants in TT system was higher than that in TR system. The same result was observed in Cd and Ni (Figures 7 and 8). Analysis of soil samples after plants harvesting (Figures 9 and 10) showed that the residual Zn and Cu values in soils were higher than other element and reached to 32, 51, 108 and 72, 63, 162 µg/g for Cu and Zn in S1, S2 and S3, respectively. In other words, application of intercropping systems significantly decreased these values. Results showed that TR system decreased Cu concentrations in soils to about 33, 51 and 109 µg/g and about 148, 52 and 59 µg/g in S1, S2 and S3 respectively for Zn. Also, TT system significantly decreased the concentration of both pollutants in used soils. The TT system decreased Cu to 72, 47, 31 for Cu and 157, 59, 67 µg/g for Zn in S1, S2 and S3 respectively; it should be noted that both pollutants beside Ni were lower in TR than in TT in all used soils. A lot of references applied the intercropping system to minimize the hazards of PTEs, for example intercropping *Cunninghamia lanceolata* with tea crops reduces the levels of Pb, Ni, Mn, and Zn in soil and tea leaves (Xue and Fei 2006). Such alteration of heavy metal absorption may be caused by the alteration in the type, amount, and function of secretions and enzymes, or inter and intra-specific competition among plants for various elements (Pivertz 2001; Silvertown and Charlesworth 2009).

**Effect of chemical and intercropping remediation on heavy metals concentration in edible parts of tomato plants**

Tables (5 and 6) represent the concentrations of the heavy metals in tomato plants after intercropping and chemical remediation systems. Chemical remediation showed priority in decreasing pollutant concentrations inside tomato fruits. Applied intercropping system to remediate the contaminated soils showed that all pollutant concentrations decreased in TT compared to TR system. Cadmium concentration values in fruits under TR system were 0.3, 0.36 and 0.45 µg/g for plants cultivated in S1, S2 and S3 respectively, while these values were decreased to 0.18, 0.22 and 0.27 µg/g in S1, S2 and S3 respectively by TT system. The same trend was observed for other elements except for Pb. Under intercropping system, the results showed that Ni concentrations in fruits were the highest values compared to other pollutants. The numerical values of Ni were 3.2, 7.9 and 10.2 µg/g in S1, S2 and S3 respectively under TR system, these values decreased to 2.8, 7.1 and 9.2 in S1, S2 and S3 respectively under TT system. The chemical analysis of used soils for different pollutants under intercropping system implies increasing of concentrations of these pollutants in S3, decreased in S2 and the lowest values were observed in S1. Under TT system, the concentration of Zn in tomato fruits cultivated in S3 was 2.76 µg/g decreased to 1.32 in S1 and decreased to 1.95 µg/g in S2 respectively.

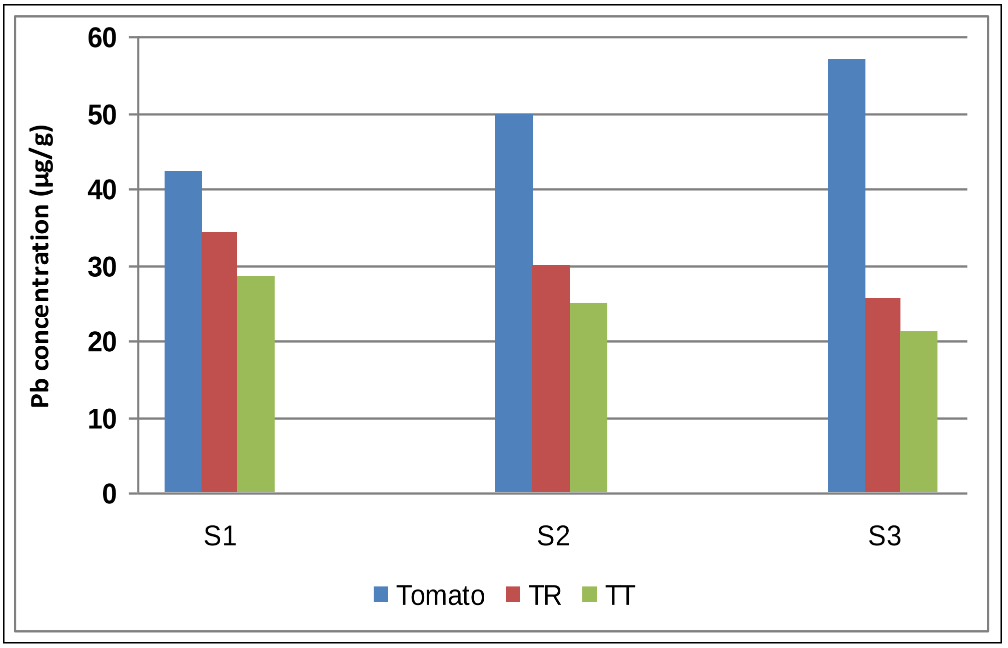


**Fig. 6. Total Pb content in the soils after application of interceroppingsystem of Tomato with Radish (TR) and Tomato with Turnip (TT)**

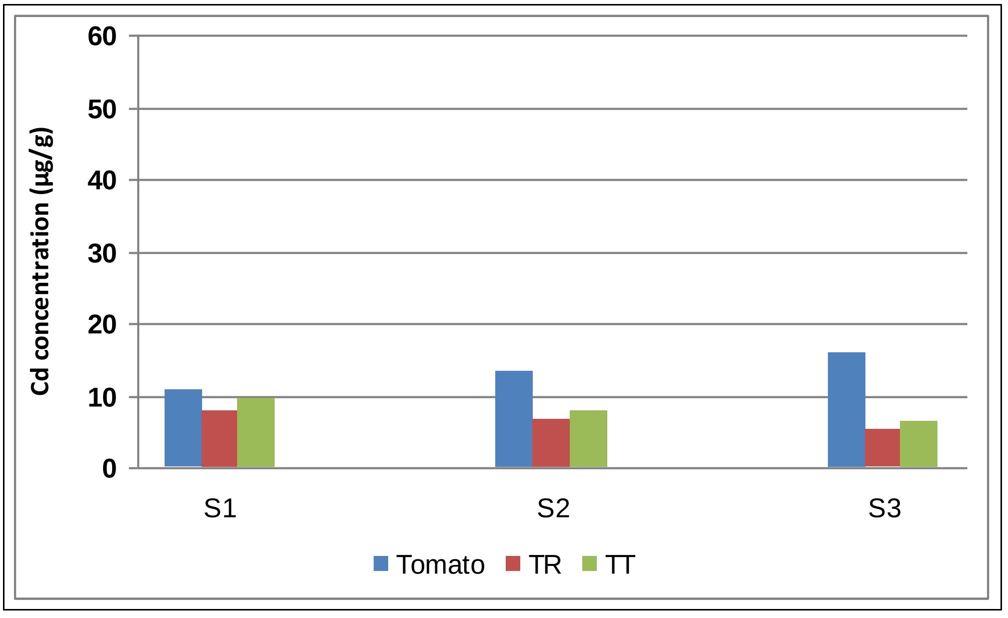


**Fig. 7. Total Cd content in the soils after application of intercropping system of Tomato with Radish (TR) and Tomato with Turnip (TT)**

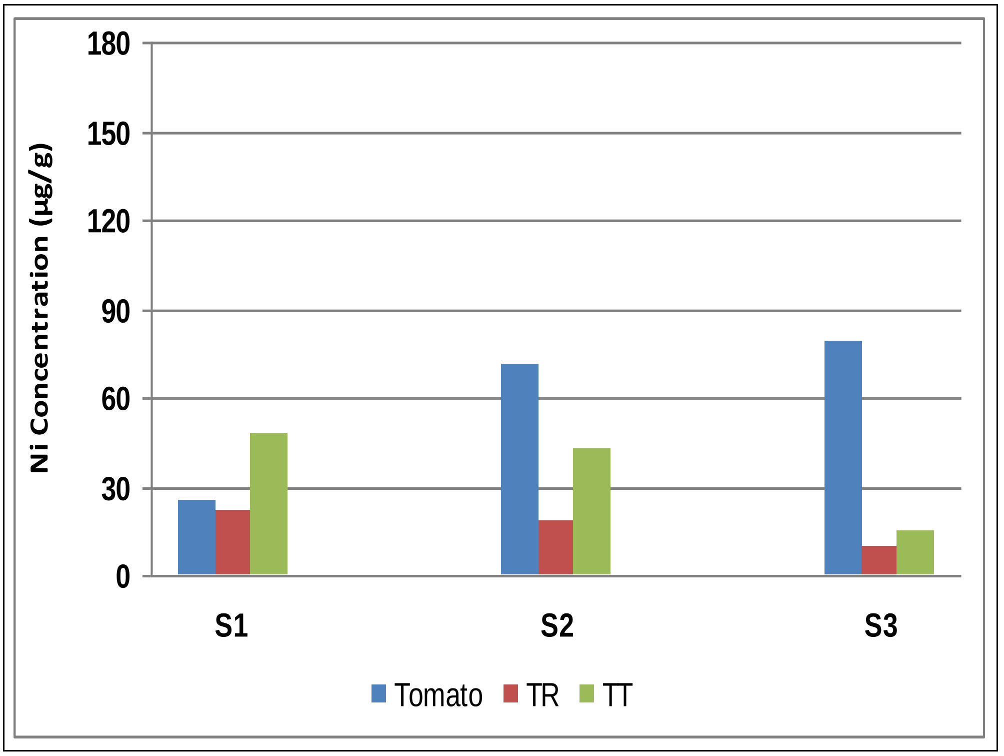


**Fig. 8. Total Ni content in the soils after application of intercropping system of Tomato with Radish (TR) and Tomato with Turnip (TT)**

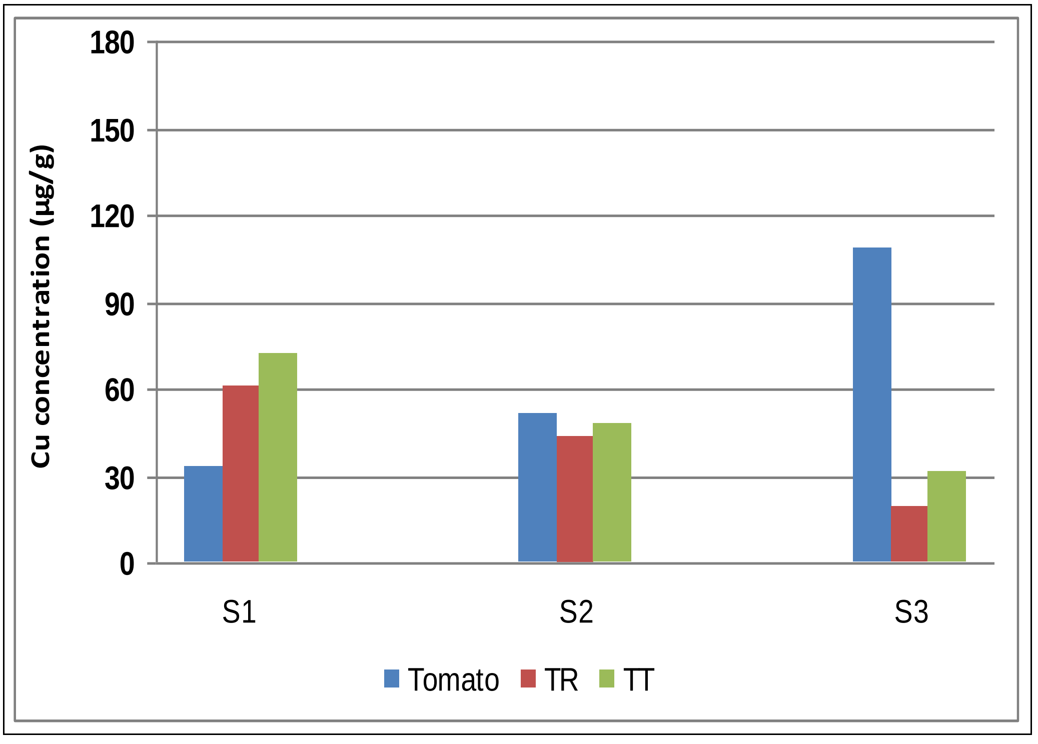


**Fig. 9. Total Cu content in the soils after application of intercropping system of Tomato with Radish (TR) and Tomato with Turnip (TT)**

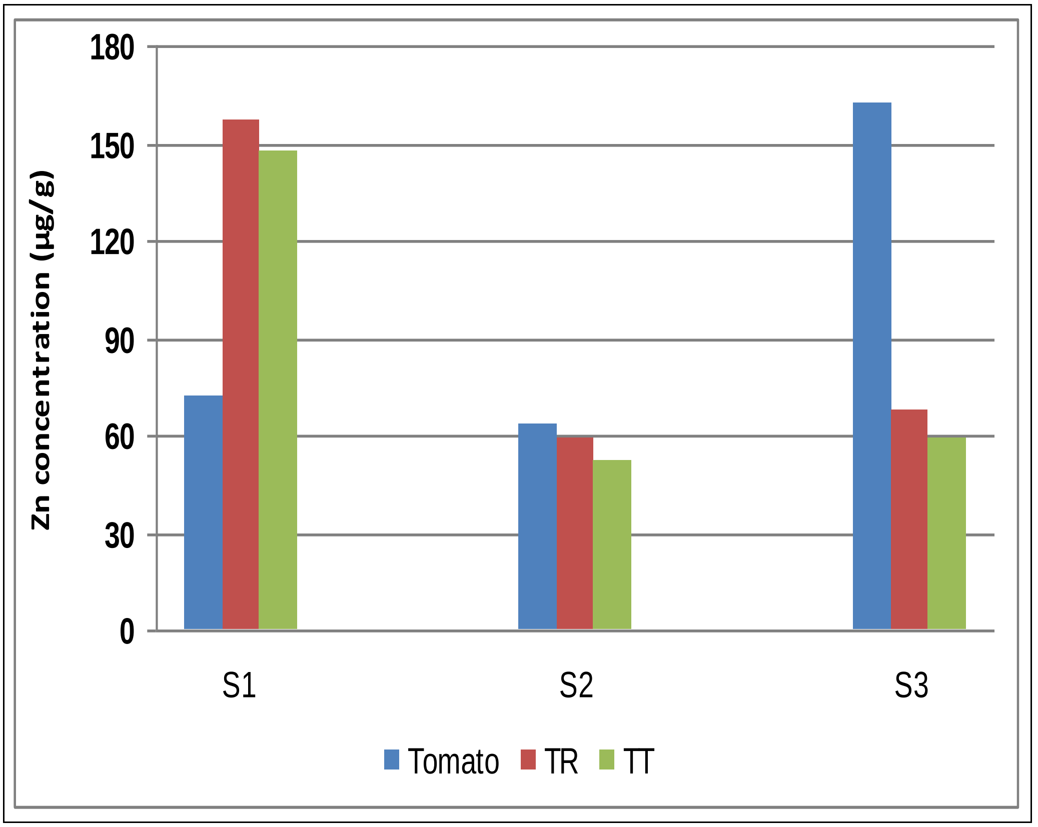


**Fig. 10. Total Zn content in the soils after application of intercropping system of Tomato with Radish (TR) and Tomato with Turnip (TT)**

**Table 5. Mean concentration of heavy metals as µg/g in the edible parts of Tomato cultivated in intercropping systems**

| Soil No. | Tomato- Radish system Fruit | Tomato-Turnip system Fruit |
|---|---|---|
| | Pb | |
| S1 | 0.12 | 0.35 |
| S2 | 0.16 | 0.38 |
| S3 | 0.23 | 0.46 |
| | Cd | |
| S1 | 0.30 | 0.18 |
| S2 | 0.36 | 0.22 |
| S3 | 0.45 | 0.27 |
| | Ni | |
| S1 | 3.16 | 2.84 |
| S2 | 7.93 | 7.14 |
| S3 | 10.2 | 9.18 |
| | Cu | |
| S1 | 5.33 | 4.8 |
| S2 | 7.28 | 6.55 |
| S3 | 7.65 | 6.89 |
| | Zn | |
| S1 | 3.44 | 1.32 |
| S2 | 3.98 | 1.95 |
| S3 | 6.34 | 2.76 |

The same trend was also observed in both TR and other pollutants. This result may due to the retention of pollutants in S3 for their highest clay content compared to other studied soils. The chemical remediation technique applied in this work showed the priority of minimizing the hazards of PTEs in tomato fruits. (Table 6) showed that the concentration of Ni was decreased in the range between 3 and 10 µg/g in TR intercropping system to the range between 1 and 8 µg/g in different soils used. Although PR fortified with PDB T1 and clay mineral used $T_2$, both significantly decreased pollutants inside the fruits, the application of T3 was the best in giving the lowest pollutants values. For example, application of T3 in polluted soils decreased Cd contents in the fruits from 0.18, 0.13 to 0.10 µg/g in S1, the corresponding values of other soils were 0.27, 0.22 decreased to 0.18 µg/g and 0.23, 0.18 decreased to 0.15 µg/g in S2 and S3 respectively, it should be mention that the same trend was observed in other elements.

**Zn equivalent values of contaminated soils treated with different techniques**

The Zinc equivalent (ZE) parameter was established to evaluate the level of contamination and the succession of remediation technique(s) in a given soil or aquatic ecosystem and it should not exceed 200 (Saber, et. al., 2012). The Zn equivalent values, which repeatedly applied to level the contamination statues of soils under the various remediation and phytoremediation treatments in the experiment are shown in Figure (11). In the current work soil quality criterion index (Zn equivalent model) was numerically calculated for the levels of PTEs toxicity according to the following equation (µg/g), the model could be written as follow:

**Zn concentration × 1 + Cu concentration × 2 + Ni concentration × 8**

A quality criterion index over 250 units indicated a risky situation necessitating remediation for better farming management (Saber et al., 2012). The calculated soil quality criterion index for the different contaminated soil samples collected from Bahr El-Baqar farms are presented in (figure 5). The calculated Zn equivalent models in the studied untreated soils were ranged between 335 and 1017; these values were extremely higher than the optimum or even critical levels (Abouziena et al., 2014).

**Effect of phytoremediation technique on ZE values**

Results in figure 11 showed that ZE values of soils remediated by phytoremediation are higher than critical value i.e. 250 (Saber et al., 2012) of the studied parameter. Numerically, tomato with turnip system (TT) decreased ZE values in S1 from 336 in control (uncultivated soil) to 280, the corresponding values of other soils were 672, 456 and 1016 decreased to 642 for S2 and S3 respectively. Cultivation of tomato with Radish (TR) significantly decreased ZE values compared to uncultivated soil (control). Results imply that ZE values of RT- system decreased from 336 in control to 269 in S1. In S2, ZE values were decreased from 672 in control to 416 as well as the corresponding values for S3 were 1016 in control decreased to 598. Although all phytoremediation plants cultivated in contaminated soils significantly decreased ZE values compared to uncultivated soils, the parameter still higher than the critical level (CL) that should not be exceeded 250. In addition, results showed a preferring of cultivating radish in contaminated soils compared to the cultivation of turnip. In all soils used cultivated with Radish, ZE values were lower than the same soils cultivated with Turnip.

**Effect of Applied Chemical Remediation Technique on ZE values**

Table 11, showed that application of rock phosphate fortified with phosphate dissolving bacteria (T1) to the contaminated soil ecosystem significantly decreased ZE value from 336, 672 and 1016 in S1, S2 and S3 uncultivated soils to 94, 186 and 190, respectively. Application of montmorillonite clay mineral (T2) decreased ZE values to 61, 152 and 169 in S1, S2 and S3, respectively. Both the applied treatments significantly decreased ZE parameter to under the critical level (CL), 250. Under these treatments, it seems reasonable to state that PTEs in the soil ecosystem goes through three chemical reactions, i.e., chemisorption of contaminants on the clay minerals, sorption on the applied solid rock phosphate surfaces and/or forming complex compounds with phosphate that was released later by the action of phosphate dissolving bacteria. The mixture of all premeditative amendments significantly decreased ZE values to the lowest ones compared to all other treatments. As shown in T3 in the figure 5, it was the best management practice in lowering ZE parameter compared to other treatments. The application the mixture treatment, the best treatment decreased ZE values from 336, 672 and 1016 in controls of S1, S2 and S3 to 61, 127. Lukman *et al.* (2013) studied residence time effects of Zn sorption on clay mineral in the presence of copper, which was added for 1 to 336 hours after Zn at a Zn/Cu molar ratio of 2. They recorded an increase in Zn sorption, especially when Cu was added for 6 to 336 hours after Zn.

They linked their findings with the initial sorption of PTE's on the surfaces of variable charge minerals that slowly released form precipitates as time goes on and/or penetrate into micro-pores of the sorbents. The same trend was observed in both cultivated and other treatments in the current work.

**Table 6. Heavy metal contents (µg/g) in the edible part of tomato plants grown in in chemical remediated soils**

| Treatment | Pb | Cd | Ni | Cu | Zn |
|---|---|---|---|---|---|
| | | | S1 | | |
| T1 | 0.18 | 0.36 | 8.16 | 6.12 | 5.07 |
| T2 | 0.13 | 0.29 | 6.34 | 5.82 | 3.18 |
| T3 | 0.10 | 0.24 | 2.53 | 4.26 | 2.75 |
| | | | S2 | | |
| T1 | 0.14 | 0.27 | 6.12 | 4.59 | 3.80 |
| T2 | 0.09 | 0.22 | 4.76 | 4.37 | 2.39 |
| T3 | 0.07 | 0.18 | 1.89 | 3.19 | 2.07 |
| | | | S3 | | |
| T1 | 0.12 | 0.23 | 5.1 | 3.83 | 3.17 |
| T2 | 0.08 | 0.18 | 3.97 | 3.64 | 1.99 |
| T3 | 0.06 | 0.15 | 1.58 | 2.67 | 1.72 |

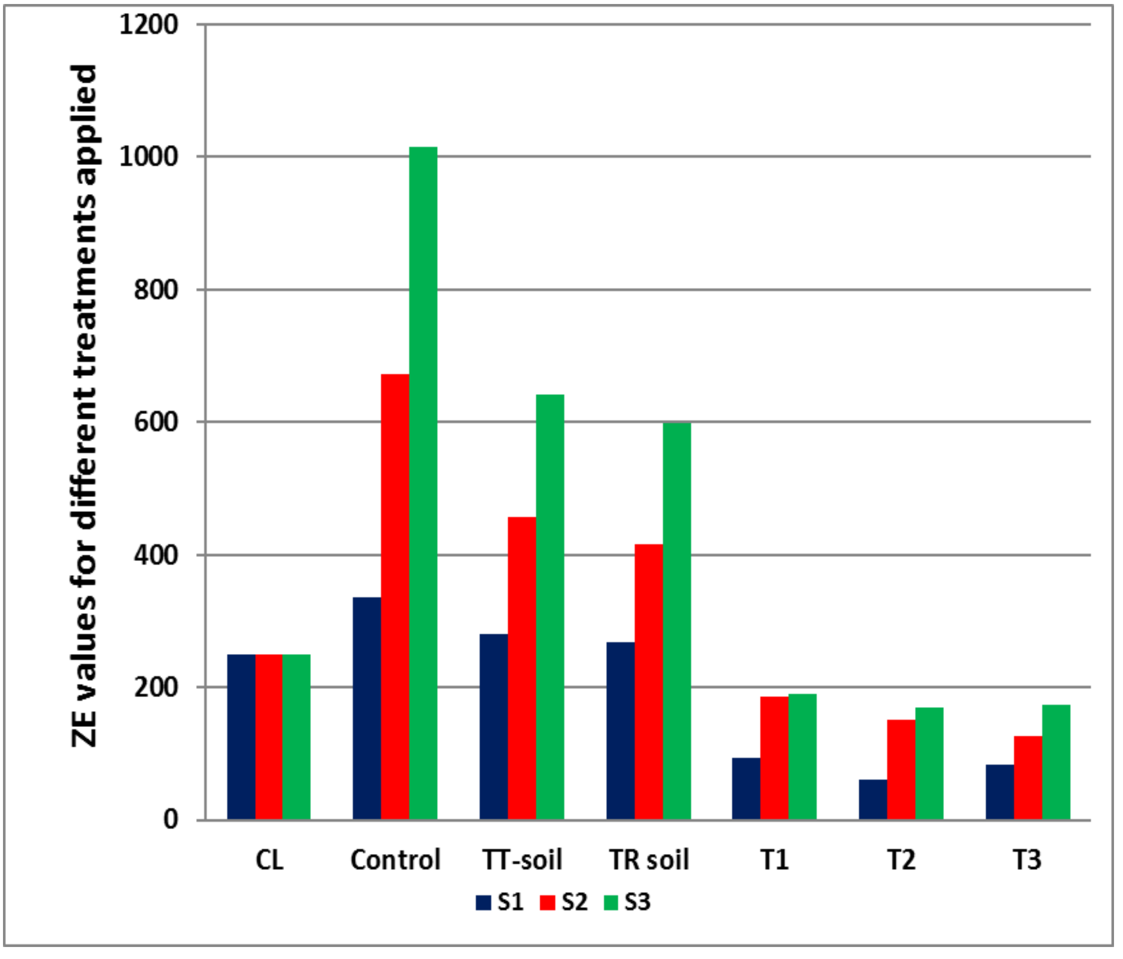


**Fig. 11. Zn equivalent (ZE) values in contaminated soils exposed to remediation with different remediate techniques, CL: critical level**

## CONCLUSION

Heavy metal contamination of ecosystem is a major environmental concern. In order to reduce the level of metal contamination, several remediation technologies have been implemented. These techniques include immobilization methods with the help of low-cost absorbent, application of some chelating agent and biology-based technique, i.e., chemical and phytoremediation. The aim of all the remediation technologies should be linked with agricultural production, food security and scale down land tenure problems. The results indicated that Chemical remediation technique was preferable to use intercropping technique. However, all techniques significantly decreased pollutants found in the tested soils.

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## الملخص العربي

# تقليل مخاطر العناصر الثقيلة فى مزارع الخضر بإستخدام تقنية المعالجة النباتية

شيرين شحاتة مريد، سحر محمد اسماعيل، علاء زغلول

تناولت هذة الدراسة إستخدام وتقييم تقنيتين لتقليل والحد من مخاطر العناصر الثقيلة الضارة (PTEs) وذلك في ثلاثة أنواع من الأراضى الملوثة من المنطقة المنزرعة حول مصرف بحر البقر وتتمثل تلك التقنيات فى: المعالجة الكيميائية والتي يمثلها أستخدام صخر الفوسفات (PR) المعامل ببكتيريا إذابة الفوسفات(PDB) وهى المعاملة الأولى، وأحد أهم معادن الطين (Montmorillonite, MCM) التى تستخدم لتقليل مخاطر تلك العناصر الضارة وهى المعاملة الثانية ومزيج من كلا المعاملتين وهى المعاملة الثالثة. تضمنت التقنيات نظم تحميل متمثلة فى أستخدام نوعين من أنواع النباتات التى لديها قدرة عالية على أمتصاص العناصر الثقيلة وهى نباتات الفجل (*Raphanus raphanistrum*) واللفت (*Brassica rapa*) والمنزرعين مع نبات الطماطم كمحصول إقتصادى زراعي وهو أحد المحاصيل الحساسة للتلوث بالعناصر الثقيلة الذى أستخدم كمؤشر لتقييم التقنيات المستخدمة لعلاج الأراضى الزراعية مختلفة القوام من رملية طينية لومية إلى طينية والتى تعانى من التلوث بالعناصر الثقيلة من مصادر متعددة وصور متنوعة لها عن طريق الرى بمياه مصرف بحر البقر. تشير النتائج المتحصل عليها إلى أن كلتا التقنيتين خفضتا معنويا تركيز الملوثات الضارة مثل Pb وCd وNi وZn وCu في الأراضى تحت الدراسة، ومع ذلك فإن تقنية المعالجة الكيميائية وبصفة خاصة الخليط بين المواد المستخدمة (المعاملة الثالثة) T3 كانت الأكثر فاعلية في تقليل مخاطر الملوثات في الأراضى تحت الدراسة. وقد نوقشت الميكانيكيات المختلفة التى تمت بين كلا من التقنيتين والعناصر الثقيلة.