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Remediation of Sanitary Wastewater containing Pb and Cd for Reuse in Irrigation

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ABSTRACT

Heavy metals found in municipal wastewater can cause many problems for human hygiene and environment. So that it should be decontaminated from heavy metals before being used in irrigation. Many materials such as shall, alginate and iron oxide were used to remove heavy metals (Pb and Cd) from municipal wastewater. The results showed that, shall had the highest efficiency than the other materials in removing Pb and Cd from polluted water. Artificial polluted water was prepared by diluting stock solution of Pb and Cd of concentration of 1000 mg/l using distilled water. The prepared Pb and Cd polluted solutions had concentrations of 5, 10 and 50 mg/l. The remedy agents were added to polluted water at ratios of 1:100, 1:1000 and 1:10000. The solutions were gently agitated to equilibrium periods of 2, 6, 12 and 24hr. Generally, the efficiency of remedy agents to remove heavy metals increased as the addition ratio increased. Percentages of Pb removed by mudstone at addition ratio of 1:1000 were 86 and 32.8% for 10 and 50 mg Pb/l, respectively, in a reaction period of 2 hrs. Regarding Cd, the removal % was 84 and 66.4 for 5 and 50 mg Cd/l, respectively, in 2 hrs. On the other hand, the Percentage of Pb removed by iron oxide and alginate were as low as 0 and 30 %, respectively. Generally for all treatments, the removal efficiency decreased as heavy metal concentration in polluted water increased. Shall was the most effective material in decontamination of heavy metals polluted water and it could be recommended to be used to decontaminate wastewater.

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Introduction

Heavy metals are ubiquitous in the environment, as a result of both natural and anthropogenic activities. Human are exposed to heavy metals through various pathways (Wilson and Pyatt, 2007). Using wastewater in irrigation, solid waste disposal, sludge applications, vehicular exhaust and industrial activities are the major sources of soil contamination with heavy metals, and an increase in metal uptake by food crops grown on such contaminated soils is often observed. In general, domestic wastewater contains substantial amounts of beneficial nutrients and toxic heavy metals, which are creating opportunities and problems for agricultural production, respectively. The most dangerous toxic elements listed by the European Economic Community on a “Black List”, were Hg and Cd, while the less dangerous substances forming the “Grey List” were Zinc, Copper, Nickel, Chromium, Lead, Selenium, Arsenic, Antimony, Molybdenum and Titanium (Bond, P. B. R., 1995). Excessive accumulation of heavy metals in agricultural soils through wastewater irrigation, may not only result in soil contamination, but also lead to elevated heavy metal uptake by crops, and thus affect food quality and safety (*Muchuweti et al ., 2006*). Heavy metals accumulation in soils and plants are of increasing concern because of the potential human health risks. Thus food chain contamination is one of the important pathways for the entry of these toxic pollutants into the human body.

Cadmium is present in wastewaters from metallurgical alloying, ceramics, electroplating, photography, pigment works, textile printing, chemical industries and lead mine drainage. The application of phosphate fertilizers or sewage sludge may increase cadmium levels in soil, which, in turn, can cause increases in cadmium levels in crops *(Gadd, G. M. and White, C., 1993)*. Cadmium is not known to have any beneficial effects, but can cause a number of adverse health effects. The U.S. department of Health and Human Services has determined that cadmium and certain cadmium compounds may reasonably be anticipated to be carcinogens. Cadmium is also one of the most toxic heavy metals and is considered nonessential for living organisms. This element is found at low concentrations in natural environments, but human activities have led to increased levels in all the continents (*Evange1ou V.P., 1998)*.The average cadmium content of sea water is about 0.1ug/1 or less (*Egyptian Environmental Affairs Agency "EEAA", 1992)*. While river water contains dissolved cadmium at concentration of < 1.1 - 13.5 ng/l Cadmium levels of up to 5 mg/kg have been reported in river and lake sediments and from 0.03 to 1 mg/kg in marine sediments *(Eckenfelder W. and Wesley Jr., 1989)*. A drinking water guideline value of 0.003 mg/l has been set for cadmium by WHO. In addition, the provisional tolerable weekly cadmium intake must not exceed 7 µg/kg of body weight *(World health organization, 1993)*.

The source of lead in wastewaters is the storage-battery manufacture (*Eckenfelder W. and Wesley Jr, .1989)*. Battery manufacturing wastewater consists mainly of H_2S0_4 solution at pH 1.2-2.5 in water which has a composition similar to tap water

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and contains soluble lead. Thereby stringent regulations are applied to wastewater effluents (*Pacyna, J. M., 1986)*. Damage to the nervous system and gastrointestinal symptoms are the main signs of lead poisoning. Lead also interferes with the formation of red blood cells, leading to anemia. Lead is especially toxic to the growing brain and can affect the behavioral development of young, even at low concentrations. For example, in polluted cities, fumes affect children's development. Lead can even pass through the placenta and thus affect a growing fetus. Organic lead compounds are fast-soluble and are more toxic than other forms *(USEPA, Environmental Protection Agency, 1986*). It was established that, a provisional tolerable weekly lead intake of 25mg/kg of body weight (equivalent to 305 mg/kg of body weight per day) for infants and children who took account of the fact that lead is a cumulative poison so that any increase in body burden of lead should be avoided *(Kocaoba S. and Akein G., 1997*). The guideline value for lead in drinking water given by WHO is 0.01 mg/l *(World Health Organization, 1984)*.

The percent study aimed to achieve an efficient non-expensive and environmentally safe method to decontaminate heavy metals (Pb and Cd) from polluted wastewater, Natural and non expensive materials, Alginit, shale and iron oxide, were used to decontaminate heavy metals polluted water to be suitable for discharging in drains and sanitary sewer system.

Materials and Methods

1- Wastewater:

Wastewater samples which was collected from three different sources including:

a- Domestic wastewater which was collected from four treatment plants. These plants are Shoubra el-Kheima, El-Birka, Zenien and Abo-Rawash.

b- Agricultural Wastewater which was collected from main drains of Bahre El-Baqare and Qualuope, and Lake of Manzala.

c- Industrial wastewater which was collected from Mac-Carpet plant, The 3rd industrial region at 10th of Ramadan, Helwan and Tebeen region that was a collection of plastic plant, iron and steel plant and coke plant , and Copper foundries in middle Cairo.

2- Artificial polluted water:

Artificial polluted water was prepared by accurate diluting of standard heavy metals solution of 1000 mg/l to known concentrations using distilled water. Lead polluted water was prepared using lead stock solution (1000 mgPb/l in 0.5 M HNO_3 as matrix). A series of standard lead solutions of 5, 10, 50 and 100 mgPb/l were prepared and used to test the ability of remedy agents in Pb removal. Cadmium polluted water was prepared using cadmium stock solution (1000 mgCd/l in 0.5 M HNO_3 as matrix). A series of standard cadmium solution of 5, 10, 20 and 50 mgCd/l were prepared and used to test the efficiency of remedy agents in Cd removal.

3- Remedy Agents:

Three remedy agents, Alginit, active iron oxide, and mudstone were used for polluted water remediation trials. These reagents are inexpensive and easily to be obtained. Besides, they could be separated easily from treated water at the end of the remediation process. These agents were selected based on their high adsorption capacity which attributed to their high surface area. The important Characteristics of the remedy agents are summarized in the following paragraphs.

Alginit

Alginit is a natural rock out of the oil shale family. It originated from fossil algae biomass and pumice, descents from the mine in Gerce, Hungary. The essential ingredients of Alginit are the high content of organic matter (19%), clay (54%) and lime content (22%). The clay is rich in montmurillionte (52%).

Mudstones

Shall is a naturally occurring material exists in many places in Egypt at different depths. It is mainly consists of clay (55%).The clay is rich in Montomorillonit. Chemical analysis showed that the mudstone contains high amount of salt, the electrical conductivity (EC) of 1:2.5 water extract equals 10.63 dS/m and pH = 7.31. Sodium was the dominate cation.

Iron Oxide (60 % Fe)

The sample of iron oxide is imported from Roseland Kazreti. The Chemical composition the iron Oxide is Fe_d (60%), Fe_o (19%), Al (0.16 mg/ kg), Zn (12.8 mg/kg) and Cu (9.94mg/kg).

4-Remediation trials

Remedy agents of Alginit, mudstone, and iron oxide, were added to heavy metals polluted water at different solid: solution ratios of 1:100, 1:1000 and 1:10000. The mixtures were then gently agitated and submit to different equilibrium periods of 1, 5 and 24 hrs. At the end of each equilibrium period, the supernatant solution was obtained by centrifuging the mixtures at 3000 rpm for 10 minutes. Concentrations of studied heavy metals (Pb and Cd) were determined for the reacted solutions using Flam Atomic Absorption Spectrophotometer (UNICAM 969, APHA, 1998). Concentrations of Pb and Cd were determined for the reacted solutions before adding the remedy agent and at the end of the reaction period. pH of the reacted solutions were measured before adding the remedy agent and at the end of different equilibrium period. All trials were done in two replicates.

5-Analytical Procedures

Concentrations of Pb and Cd in polluted water as well as treated ones were determined using Atomic Absorption Spectrophotometer (UNICAM 969) (APHA, 1998). The quality standards of the determinations are listed in Table (1). The instrument was adjusted before each run using a series of standard solutions of concentrations identical to the measured samples.

The Electrical Conductivity (EC) of the reacted solutions was measured using digital YSIEC meter (model 35).

pH was measured using digital Orion pH meter (model 420A) in which the pH was measured as potential difference between Glass Electrode and Calomel Electrode. The pH-meter instrument was adjusted before measuring using three buffer solutions of pH 4, 7 and 9.

Results and Discussion

The concentration of heavy metals in wastewater collected from different sources (domestic, agricultural and industrial) are presented in Table (1). These concentrations are relatively low due to dilution effect since the initial wastewater has been mixed with other sources of water. That is besides, most of heavy metals are slightly soluble and tend to be adsorbed and precipitated on the surface of solid particles exist in wastewater.

Mudstones:

Efficiency of mudstone to remove Pb and Cd from solutions contain various concentrations of 5, 10 and 50 mg/l were examined using different addition ratios of shall: polluted solution, at different reaction periods varying between 1 to 24 hrs.

The results showed, low addition ratio (shall: heavy metal polluted solution) of 1: 10000 was effective in removing either Pb or Cd in solutions containing low concentration of 5 mg/l. As the addition ratio increased to 1:1000, the removal efficiency slightly increased. At addition ratio of 1:1000, shall was able to reduce Pb and Cd concentration from 5 to be 1.14 and 0.34 mg/l, respectively, in one hr reaction time (table 2). At addition ratio of 1: 100, the efficiency greatly increased (table 3). Mudstones successfully reduced the initial Pb concentration of 5 and 10 to be 0.4 and 0.7 mg/l, in a reaction period of 1 hr which were lower than the permissible level (5 mg/l) for irrigation water. The corresponding values for Cd were 0.22 and 0.74 mg/l, which were little higher than the permissible level (0.01mg/l) for irrigation water.

Mudstone proved efficiency to remediate higher concentration of 20 mg Cd /l and at high addition ratio of 1:100. It reduces Cd concentration from 20 to 1.5 mg /l in a matter of 2 hours reaction time. High reaction time of 24 hrs is not recommended for mudstones due to the release of adsorbed Pb and Cd, (table 2 and 3).

Alginit

Regarding Pb, the lowest addition ratios of 1:10000 was not effective even for low concentration of 5 mg Pb/l. Addition ratio of 1:1000 (Table 4) was effective only for relatively low concentration of 5mg /l, in which it significantly reduced to 0.52 mg/l in a reaction time of 24 hrs. The efficiency of Alginit in remediation of Pb polluted water increased as the addition ratio increased. Addition ratio of 1:100 significantly eliminates Pb from solutions contain 5 and 10 mg Pb/l in a reaction time of 24 hrs, Pb concentration in the previous solutions reduced to be 0.21 and 0.46 mg/l, respectively. For solution of 50 mg Pb /l, addition ratio of 1:100 (table 5) was able only to reduce Pb to level (6.5 mg/l) nearby the permissible level.

Regarding Cd, low addition ratios of 1:10000 and 1:1000, showed low efficiency to remove Cd from polluted water. However, high addition ratio of 1:100 proved high efficiency in eliminating Cd from polluted water. It reduces the initial Cd concentrations from 5 and 10 to 0.21and 1.57 mg/l, respectively. The results show that, removal of Cd by Alginit was time independent since the changes in pollutant concentrations with time was insignificant. Based on the Cd permissible level in irrigation water (0.1 mg/l), Alginit showed low efficiency for removing Cd. It works only with low Cd concentration and high addition ratio of 1:100.

Iron oxide

Iron oxide proved efficiency in removing Pb from polluted water only at high addition ratio of 1:100. This efficiency was extended even for high Pb concentration of 50 mg/l. The efficiency of iron oxide increased as the reaction time increased. In a reaction time of 24 hrs, it eliminates Pb in solutions of initial concentration of 5 and 10, in which the concentration reduced to be Nil (Figure 1 and 2). Also, it reduced the concentration of 50 mg Pb/l to be 1.67 mg/l (Figure 3) which is lower than the permissible level (5 mg/l) for irrigation water. Addition ratio of 1:1000 works well with low Pb concentration of 5 mg /l, which declined to be 0.67 mg Pb/l. As Pb concentration increased to be 10 mg/l, the efficiency dropped, in which the equilibrium concentration dropped to be 3.5 mg/l after 24. However, the lowest addition ratio of 1:10000 was ineffective in removing the metal even from water polluted with low Pb concentrations of 5 mg/l.

The efficiency of Fe-oxide in removing Cd was time dependent. As the reaction time increased, Cd concentration in the equilibrium solution decreased. High addition ratio of 1:100 was the most effective in removing Cd. Cadmium removal efficiency sustains for Cd concentration up to 10 mg/l. Initial concentration of 5 and 10 mg Cd/l (Fig.1 and 2) declined to be 0.1 and 0.4 mg/l, respectively, after 24 hrs at addition ratio of 1:100. Cd concentration (0.1 and 0.4 mg/l) in the treated solution were identical and little higher, respectively, than the permissible level (0.01 mg/l).

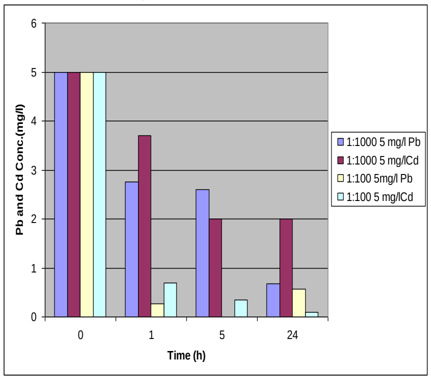

Fig.(1): Concentrations of Pb and Cd in water polluted with 5 mg/l after being treated with iron oxide at addition ratios of 1:1000 and 1:100 for reaction times of 1, 5 and 24 hrs.

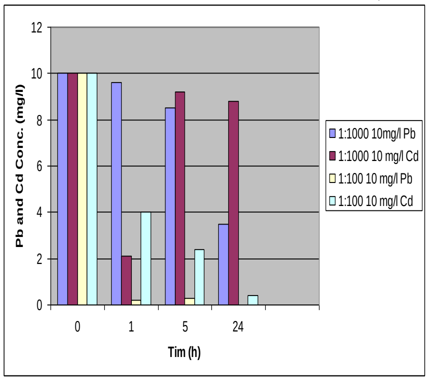

Fig.(2): Concentrations of Pb and Cd in water polluted with 10 mg/l after being treated with iron oxide at different addition ratios 1:1000 and 1:100 for reaction times of 1, 5 and 24 hrs.

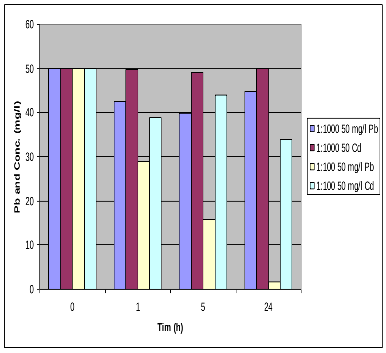

Fig. (3): Concentrations of Pb and Cd in water polluted with 50 mg/l after being treated with iron oxide at different addition ratios of 1:1000 and 1:100 for reaction times of 1, 5 and 24 hrs.

Table (1). Concentrations of heavy metals in collected wastewater.

Wastewater Sources	Fe	Mn	Cd	Co	Ni	Pb	Cr	Cu	Zn
El-Birka,	0.56	0.06	0.003	0.004	Nd	0.02	0.01	0.003	0.06
Shoubra el-Kheima,	0.65	0.20	0.003	0.002	Nd	0.11	0.001	0.016	0.14
Zenien	0.77	0.15	0.006	nd	Nd	0.062	nd	0.04	0.11
Abo-Rawash.	0.53	0.08	Nd	nd	0.003	0.037	nd	0.05	0.10
Bahre El-Baqare	0.54	0.31	0.005	0.006	nd	0.05	0.013	0.04	0.06
Lake of Manzala.	0.61	0.30	0.004	0.002	nd	0.034	0.006	0.04	0.06

Table (2) Concentration of Pb and Cd in polluted water before and after being treated with mudstone at addition ratio of 1:1000 for different reaction times

Concentration(mg/l)	5			10			50		
Time (hr)	1	5	24	1	5	24	1	5	24
Pb	1.14	1.14	1.68	2.70	2.60	4.40	32.70	32.70	39.77
Cd	0.34	1.11	1.43	5.00	3.00	4.40	41.50	28.80	50

Table (3) Concentration of Pb and Cd in polluted water before and after being treated with mudstone at addition ratio of 1:100 for different reaction times.

Concentration (mg/l)	5			10			50		
Time (hr)	1	5	24	1	5	24	1	5	24
Pb	0.40	0.40	0.20	0.70	0.51	0.98	14.50	15.30	21.59
Cd	0.22	0.21	0.07	0.74	0.77	0.68	17.80	13.60	14.00

Table (4) Concentration of Pb and Cd in polluted water before and after being treated with Alginit at addition ratio of 1:1000 for different reaction times

Concentration(mg/l)	5			10			50		
Time (hr)	1	5	24	1	5	24	1	5	24
Pb	1.85	1.2	0.52	9.01	8.10	9.68	40.20	39.70	44.55
Cd	2.80	4.40	3.20	9.10	8.00	5.20	50	48.9	50

Table (5) Concentration of Pb and Cd in polluted water before and after being treated with Alginit at addition ratio of 1:100 for different reaction times.

Concentration (mg/l)	5			10			50		
Time (hr)	1	5	24	1	5	24	1	5	24
Pb	0.04	0.21	0.21	0.4	0.13	0.46	19.80	17.9	6.50
Cd	0.29	0.23	0.21	2	1.70	1.57	47.40	39.9	30.00

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