# Evaluation of the Effect of Reverse Current and Advective Flow on Heavy Metals Removal from Municipal Wastewater Sludge

## Hazem I. Saleh<sup>1</sup>

<sup>1</sup>Civil Engineering Department, Menoufia Univ. Egypt <sup>1</sup>

### ABSTRACT

Heavy metals removal from dewatered municipal wastewater sludge under the effect of electric potential and advection flow was investigated. The application of these processes resulted in 24%, 10%, and 8.2% removals of Ni, Cr, and Fe respectively with higher removal at the anodic site. Lower removals for Zn, Cu, and Pb (1.8%, 3.5%, and 3.2% respectively) with more efficient removal at the cathodic site were obtained. Since metals of high concentrations in the sludge were removed mainly at the anodic site, a reverse charge concept was implemented by interchanging the cathode and anode of the electrokinetic cell alternatively. Results showed an average removal efficiency for Cu and Ni removal compared with hose obtained from the first investigation. Higher efficiency for Zn removal was observed, but lower efficiency for Cr, Fe, and Pb were obtained.

Keywords: wastewater sludge, heavy metals removal; electric potential, advection, land application.

#### Introduction

Heavy metals can reach municipal wastewater sludge via discharge of uncontrolled industrial effluent to public sewers thus increasing the metal content of sludge and imposing a problem for proper sludge management. While efforts are made to prevent heavy metals discharge through controlled industrial effluent discharge and installation of pretreatment units at generation sources, alternative solutions should be considered for metals removal from the actually generated municipal wastewater sludge.

Manuscript received from: Dr. Hazem I. Saleh at: 26 /4 / 1998 Accepted at: 19 / 7 / 98 Engineering research bulletin, Vol. 21 No.3 1998 Menoufiya University, Faculty of Engineering Shebin El-Kom, Egypt, ISSN. 1110 - 1180 Land application of the sludge for use in agricultural purposes is one of the vital alternatives considered for sludge management. Since heavy metals tend to accumulate in the food chain, consequent health hazards can be minimized by setting limits for heavy metals cumulative concentrations applied to agricultural land. Therefore, metals concentration in raw sludge will dictate its long term use for agricultural purposes and additional measures are required for sludge metal decontamination to further prolong its use. Heavy metals such as zinc, copper, nickel, lead, and chromium are of concern in sludge application for agricultural purposes due to their phytotoxicity effect. Apart from heavy metals, pathogens are also of concern in agricultural application, and treatment methods as lime stabilization or composting are applied to reduce the number of pathogens in the sludge (U.S. EPA, 1992). Other alternatives as incineration, landfilling, or dedicated land disposal have less constraints on sludge metal content.

Several processes for heavy metals removal from the sludge were investigated including lowering the sludge pH to increase metals solubility and mobility (Fristoe and Nelson, 1983), and metals leaching biologically using indigenous sulfur-oxidizing bacteria present in the sludge (Sreekrishnan et al., 1993; Tyagi et al., 1994). Heavy metals partitioning in municipal sludge was previously investigated and the possible forms of metals combination in sludge were summarized in the following: (a) in mineral grains exchangeable cations on clays, (b) discrete crystals or grains of secondary precipitates or in coatings, (c) as intra-cellular or extra-cellular uptake by bacteria, (d) organic complexes on colloid, and (e) complex ions (MacNicol and Beckett, 1989). Greater fraction of trace metals is held in both the larger fraction of sludge having low density and high organic content and as precipitates of grains of higher density (Ravishankar et al., 1994). While the movement of metals can be reduced by maintaining the soil at pH 6.5 or above, metal availability to crops constitutes a problem that may shorten municipal sludge application in agricultural purposes due to high metal concentrations. Table 1 shows the recommended cumulative limits for metals of major concern applied to agricultural crop land (U.S. EPA, 1983).

The application of electric potential is currently used for site decontamination from heavy metals. In highly buffered systems without pH control at the electrodes, strong acid strong base interactions govern, and sharp pH fronts are typical. Advection, electrical conduction, and diffusion also affect metal ions mobility, species transport is influenced by both the electric field and the pH that affect metal solubility (Alshawabkeh and Acar, 1996; Eykholt and Daniel, 1994).

Electric potential application to municipal wastewater sludge directly affects heavy metals solubility and mobility through a low pH region around the anode and increased cations attraction towards the cathode. The use of hydraulic head may also affect the rate of effluent collection, ions mobility and metals removal. In this study, heavy metal removal from the sludge was investigated under the effect of electric potential with and without the use of a hydraulic head. Furthermore, the effect of reversing the electric current by cyclic alteration of the poles on metal removal was also studied.

#### Materials and Methods

Dewatered municipal wastewater sludge was collected from the City of Montreal wastewater treatment plant applying physico-chemical treatment using ferric chloride and Percol 902 polymer from Allied Colloids for the sedimentation process. Sludge is then mechanically dewatered using Percol 757 from the same manufacturer. Sludge cake samples were preserved at 4°C, heavy metals concentration in the sludge are shown in Table<sup>1</sup> 2. Approximately 200 grams of sludge cake were filled in the cell, air entrapment<sup>2</sup> was minimized, by slightly compacting the cake. The direct electric current was applied to the sludge using an electric DC power supply TES 6230 connected to both the anode and cathode of a plexiglass cylindrical cell measuring 50 mm in diameter and 75 mm in height. A constant voltage of 26.5V was applied, stainless steel discs measuring 50 mm in diameter were fixed at the two ends of the cell acting as the anode and cathode of the system.

The use of a hydraulic head to accelerate ions mobility through the system was also investigated, two different approaches were adapted in the experimental investigation. In the first approach, successive effluent weres collected from both the anode and cathode using low and high hydraulic heads via introducing distilled water at the centre of the cell. Distilled water was used to compensate for the effluent collected at both the anode and the cathode. The low head (150 mm from the centre line of the cell) was used to ensure continuous feed of distilled water to the cell and to prevent gases produced by electrolysis reactions from backing up in the water feed line. Using a low hydraulic head allows the effect of electric potential to be dominant. This was considered as Phase Eduring which 160 ml of effluent were collected at each of the anode and cathode within six hours. These effluents were designated as A(I) and C(I). When the system was accelerated using a hydraulic head of 500 mm, i.e., Phase IL approximately three times the effluent of Phase I were collected within 190 minutes. These effluents were designated as A(II) and C(II) for anodic and cathodic effluents respectively.

Chemical analysis showed that some metals tend to be removed from the system through the anodic site, others from the cathodic site under the .1:1

combined effect of low anodic pH, cathodic attraction, and advection. Since metals of higher concentration in the sludge were removed from the anodic site, the second approach was introduced where the poles of the direct electric current were alternated at fixed time intervals, under the same hydraulic head used in Phase I. Only anodic effluents were collected from anode (A). After 15 minutes (time of one cycle), the poles were switched thus the former cathode is now the new anode (B) through which effluent was collected. These cycles were repeated to the end of the experiment. batches of anodic effluent collected from (A) and (B) each eight cycles (two hours) and were termed (A-15) and (B-15), referring to 15 minutes cycle, then analyzed for heavy metal contents. Time cycle interval was then raised to 30 minutes in a new experimental run and batches of effluents were collected also each eight cycles (four hours) and termed (A-30) and (B-30). Metal concentrations, variations in effluent pH and electrical conductivity were measured for the collected batches.

Heavy metal concentrations in raw sludge were determined after digesting the sludge using sulfuric acid and hydrogen peroxide. In the digestion procedure, 4.0 ml of sulfuric acid were added to 0.5 g of sludge in the digestion flask, and heated at 440°C for two to three minutes, then 10 ml hydrogen peroxide were added and the solution was left to cool for at least 30 minutes. Distilled water was added after cooling to dilute the digest to 100 ml, metal ions concentration in the digest and the effluent were determined using Perkin Elmer atomic absorption spectrophotometer.

#### **Results and Discussion**

#### First Approach: Continuous Current Application

When applying electric potential to the sludge and allowing effluent collection from both anode and cathode, various conditions such as cathodic attraction for cations, low pH at the anodic side, and high pH at the cathodic side were developed. Effluent pH is important in reflecting possible mobility of heavy metals as their solubility will be affected. The sharp difference in effluent pH (Fig. 1) can be attributed mainly to pore fluid hydrolysis, when the flow was accelerated using a hydraulic head, less acidic and basic values were obtained.

A clear change in metal removal pattern between phases was observed for Cr, Fe, Pb, and Cu removal (Figs. 2 to 4), Ni and Zn removals were not altered due to phase transition. Therefore, where Ni and Zn concentration are of concern, the use of hydraulic head is beneficial in achieving the same rate of metal removal at shorter operating time. In addition, Ni removal increased at the cathode under the effect of advective flow. Lead and copper were similar in their removal pattern mainly from the cathode. Chromium was highly removed from the anodic side mainly under the effect of low pH. Removal rate decreased as pH increased in Phase II, while removal at the cathode was low due to high pH. Iron removal was similar to that of Chromium at the anode while removal rate at the cathode increased in Phase II due to advection. Metals removal from the anode with the application of electric current is not uncommon as denoted in previous experimental studies where cations that should have been transported to the cathode compartment were found in the anode compartment (Alshawabkeh and Acar, 1996; Eykholt and Daniel, 1994).

Under the applied experimental conditions, combined effect of osmotic and advective flows through the sludge led to high removal of Ni, Cr, and Fe from the anodic side, while Zn, Cu, and Pb were efficiently removed from the cathodic side. Metals removal from the anode is justified by the low pH developed there and to the initially high concentration of Cr and Fe in the sludge. While Ni concentration was not high compared with the previous two metals, it showed a high tendency for solubility from the sludge and mobility under acidic conditions (Mohamed and Saleh, 1997). Metals removed mainly from the cathode are usually encountered in the fine particulates of sludge. Entrapment of these particulates with the generated gas and the effluent, besides the effect of cathodic attraction may all contribute to the high removal at the cathodic site.

In evaluating the effect of advection on metal removal, the percent of total removal in each phase was plotted for each metal (Figs. 5 and 6) taking into consideration that the effluent in Phase II was three times that in Phase I and was collected within 27% of the time elapsed in Phase I. Comparison was made based on the total volume collected in both phases and can otherwise be based on time basis and/or unit volume collected. Removals of Cr, Fe, Cu, and Pb were higher under electroosmotic effect, while advective flow considerably decreased Cu and Pb rates of removal at the cathode. On the other hand, advective flow increased Ni and Fe removals from the cathode: Total removals of metals from the sludge is shown in Fig. 7, removal efficiencies were 24%, 10%, 8.2%, 1.8%, 3.5%, and 3.2% for Ni, Cr, Fe, Zn, Cu, and Pb respectively.

#### Second Approach: Reverse Current

As metals of higher concentration in the sludge were removed from the anode, the second approach was introduced through alternating the current poles and collecting batches of anodic effluent each eight cycles at two different time cycle intervals of 15 and 30 minutes, i.e., collecting batches each two hours and four hours respectively. Variation in anodic effluent pH is shown in Fig. 8 from which the following can be observed: (a) in the 15 minutes cycle run, pH at both sites fluctuated over acidic and basic values, (b) in the 30 minutes cycle run, enough time allowed the first anode A to behave as the permanent anode with effluent pH<7.0, (c) the second anode B located at the other side of the cell acted as the permanent cathode with effluent pH>7.0, and (d) pH values were less acidic and basic pH values than that of A(I) and C(I).

The results obtained for heavy metal removals using the second approach, were compared with those obtained from Phase I investigated under the same conditions. Removal of Cu, and Ni are shown in Fig. 9 and 10, the removal was confined between the higher and lower rates of the continuous system. Zn showed higher removal in the 15 minutes cycle run (Fig. 11), removal of Fe, Cr, and Pb were closer to their lower concentrations in the continuous system and were not reported. The continuous system was thus considered to have an overall higher efficiency for heavy metals removal over the system of alternated poles.

The study investigated the efficiency of combined electric and hydraulic processes in heavy metals removal, the operating parameters at each run were arbitrarily chosen to be within practical ranges dealing with the daily generated sludge. These parameters, i.e. electric potential, hydraulic head, time, can be changed or optimized if a system is to be implemented for metal removal from sludge. Higher percentage of removal can be achieved upon prolonged application of the process, sludge characteristics, level of heavy metals contamination, and the suggested process for sludge management shall all be considered.

#### Conclusions

The application of an electric current and a hydraulic head to dewatered wastewater sludge resulted in high removals of Ni, Cr, and Fe mainly at the anodic site under the effect of low pH, lower removals of Zn, Cu, and Pb at the cathodic site were obtained. Total removal of Ni, Cr, Fe, Zn, Cu, and Pb were 24%, 10%, 8.2%, 1.8%, 3.5%, and 3.2% respectively. Advective flow helped the removal of Zn and Ni at the same rate within a shorter period of time, it decreased the rate of Cr and Fe removals from the anodic side due to higher pH of effluent, and increased the rate of Ni, Fe, and Zn removals from the cathodic side by overcoming the effect of high pH at the cathode that may hinder metal mobility. Alternating poles approach resulted in Ni, Cu, and Zn removals confined between the extreme values of removal in the continuous process, while low removals for Cr, Fe, and Pb were obtained.

#### References

Alshawabkeh, A.N., and Acar, Y.B. (1996) Electrokinetic Remediation. II-Theoretical Model, Journal of Geotechnical Engineering, Vol. 122, No. 3, March 1996.

- Caldwell, R.J., and Anderson, B.C. (1993) Alkaline Solubilization of Wastewater Sludges: Fate of Trace Metals, 1993 Joint CSCE-ASCE National Conference on Environmental Engineering, pp. 429 - 436.
- Eykholt, G.R., and Daniel, D.E. (1994) Impact of System Chemistry on Electroosmosis in Contaminated Soil, Journal of Geotechnical Engineering, Vol. 120, No. 5, May 1994.
- Fristoe B.R., and Nelson P.O. (1983) Equilibrium Chemical Modelling of Heavy Metals in Activated Sludge. Wat. Res. 17, pp. 771-778.
- MacNicol R.D., and Beckett P.H: (1989) The distribution of heavy metals between the principal components of digested sewage sludge, Wat. Res. V. 23, No.2, pp. 199-206.
- Mohamed, A.M.O., and Saleh, H.I., (1997) "Heavy Metals Removal from Municipal Wastewater Sludge for Land Application Management" submitted Geoenvironmental Engineering Conference, Univ. of Wales Cardiff, UK., Sep. 16-18, 1997.
- Ravishankar B.R., Auclair J.-C., and Tyagi R.D. (1994) Partitioning of heavy metals in some Quebec municipal sludges, Water Poll. Res. J. Canada, Vol. 29, No. 4, pp. 457-470.
- Sreekrishnan T.R., Tyagi R.D., and Campbell P.G.C. (1993) Kinetics of heavy metals bioleaching from sewage sludge - I. Effects of process parameters. Wat. Res. vol. 27, No. 11, pp. 1941-1651, 1993.
- Tyagi R.D., Sreekrishnan T.T., Blais J.F., and Campbell P.G.C. (1994) Kinetics of heavy metal bioleaching from sewage sludge - III. Temperature effect, Wat. Res. Vol. 28, No. 11, pp. 2367-2375.
- US. EPA. (1983) Process Design Manual: Land application of municipal sludge, Municipal Environmental Research Laboratory, EPA-625/1-83-016.
- U.S. EPA. (1987) Design Manual: Dewatering Municipal Wastewater Sludges, Office of Research and Development Washington DC 20460, EPA/625/1-87/014, p. 193.

,	Soil Cation	Exchange Capacity, me	<u>:q/100g</u>
Metal	<5	5 to 15 (kg/ha)	>15
Pb	560	1120	2240
Zn	280	560	1120
Cu	140	280	560
Ni	140	280	560

Table IV Recommended Cumulative Limits for Metals Applied to Agricultural Land

Source: U.S. EPA, 1983.

Table v Studge Heavy Metals Concentra
---------------------------------------



Figure 7 pH variation of anodic and cathodic effluent



Figure 8 Electroosmotic and advective effect on lead and copper removal





21







Figure 12 Percent of total removal for metals efficiently removed at cathode



Figure 13 Total removal of heavy metals from wastewater sludge





Figure 14 pH variation in anodic effluent at various reversed current cycles



Figure 15 Nickel cumulative removal

÷



Figure 16 Copper cumulative removal



Figure 17 Zinc cumulative removal

25

# تقييم تأثير تبادل التيار الكهربي وسريان المياه على إزالة المعادن الثقيلة من الحمأة

د. حازم إبراهيم صىالح قسم الهندسة المدنية كلية الهندسة جامعة المنوفية

ملخص

تم دراسة إز الة المعادن الثقيلة من الحمأة تحت تأثير التيار الكهربى المستمر وسريان المياه خلال الحمأة بغرض الإسراع من عملية الإز الة. وقد أسفرت النتائج عن نسب إز الة للنيكل والكروم والحديد ٢٤٪، ١٠٪، و٢٨٪ على التوالى وكانت نسبة الإز الة الأكبر من جهة القطب الموجب (الأتود). وقد تم الحصول على نسب أقل لإز الة الزنك والنحاس والرصاص (١.٨٪، ٣٥٠٪، و٣٣٠٪ على التوالى) بنسب إز الة أكبر من جهة القطب السالب (الكاثود). وقد تم إجراء تجارب تكميلية وذلك بعكس أقطاب التيار القطب السالب (الكاثود). وقد تم إجراء تجارب تكميلية وذلك بعكس أقطاب التيار المستمر لإختبار مدى تأثير ذلك على إذابة المعادن الثقيلة تحت تأثير اختلاف قيم الأس الهيدروجينى. وقد أسفرت النتائج عن نسب إز الة للنحاس والنيكل مقاربة لما تم الحصول عليه من النتائج فى مرحلة التجارب الأولى، مع زيادة فى نسب إز الة الزنك ونقص فى نسب إز الة الكروم والحديد والرصاص.