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Mobility and transport of heavy metals in polluted soil environment

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INTRODUCTION

Heavy Metals are defined as elements in the periodic table having atomic number more than 20 or densities more than 5g/cm^3 generally excluding alkali metals and alkaline earth metals. The environmental problems with heavy metals are that they as elements are undestroyable and the most of them have toxic effects on living organisms when exceeding a certain concentration. Furthermore, some heavy metals are being subjected to bioaccumulation and may pose a risk to human health when transferred to the food chain. Soils, whether in urban or agricultural areas represent a major sink for metals released into the environment from a wide variety of anthropogenic sources (Niragu, 1991).

Once in soil, some of these metals would be persistent because of their fairly immobile nature. Other metals however would be more mobile therefore the potential of transfer either through soil profile down to ground water aquifer or via plant - root uptake (bio available) is likely. When the food chain is concerned, one has to take into account the mobility and the bio availability of metals, because plant uptake of metals parallels the bio available fractions of the metals in soil.

In most soil environment sorption is the dominating speciation process and thus the largest fraction of heavy metal in a soil is associated with the solid phase of that soil. Pollution problem arise when heavy metals are mobilized into the soil solution and taken up by plants or transported to the surface/ground water. The properties of the soil are thus very important in the attenuation of heavy metals in the environment. The solubility of heavy metals in soil is controlled by reactions with solid phases. Once sewage sludge is applied to soil, the heavy metal species undergo several possible fates including

(*i*) Adsorption/desorption reactions
(*ii*) Precipitation/dissolution reaction
(*iii*) Plant uptake and
(*iv*) a possible mobility thro' soil profile

SOURCES OF HEAVY METALS IN SOIL

Arsenic (As). *It is poisonous and is used in herbicides, cattle and sheep dips and insecticides. Also as a desiccant for cotton crop to facilitate the mechanical harvesting of the crop.*

Cadmium (Cd). *Soil contamination occurs by the addition of phosphatic fertilizers. (Containing 2-200 mg Cd/kg) domestic and sewage sludge, wear of automobile tyres, lubricants and mining and metallurgical activities.*

Chromium (Cr). *Wastewater and sludge from dyeing and tanning industries are the major sources of chromium pollution to the environment.*

Lead (Pb) . *Major sources of Pb pollution are exhaust gases of petrol engines, which account for nearly 80% of the total Pb in the air. Soils located near Pb mines may contain high as 0.5% Pb content. Apart from minerals, sources of Pb are pesticides, fertilizer impurities, emissions from mining and smelting operations, an atmospheric fall out from the combustion of fossil fuels.*

Mercury (Hg). *Major contaminating sources of Hg are: Hg based funcides, sewerge sludge and atmospheric fall out resulting from combustion of fossil fuels and industrial processes.*

Nickel (Ni). *Sources of Ni pollution, are: metal refining, smelting, burning of coal and industrial sewage sludge.*

Table 1 : The heavy metal status of the sewage irrigated soils of Coimbatore.

Metals (mg kg^{-1})	Surface	Sub Surface
Cr	1.7 to 5.7	1.2 to 3.7
Cd	1.05 to 4.1	0.35 to 3.15
Pb	2.40 to 4.72	2.40 to 6.95
Ni	6.50 to 13.3	7.50 to 15.8

–Sivagami et al., (2002)

Perusal of mean data on heavy metal status indicated that all the metals were within the safer limit with the values of 3.51 and 3.25 mg kg^{-1} for Pb, 2.39 and 1.07 mg kg^{-1} for Cd, 3.52 and 2.31 mg kg^{-1} for Cr and 8.76 to 11.24 mg kg^{-1} for Ni in surface and subsurface soil respectively.

Table 2 : Speciation of heavy metals in the Sewage bio solid.

Fractions	Concentration of trace elements/heavy metals (mg kg^{-1})							
	Zn	Fe	Mn	Cu	Cd	Cr	Pb	Ni
Water soluble	0.95	29.5	6.75	5.25	0.50	9.25	8.55	10.3
Exchangeable	24	48.3	38	8.62	2.92	38.5	52.5	28.5
Organically bound	175	334	113	560	22.0	260	196	280
Carbonate	282	1744	296	330	133	161	533	268
Residual	1810	31800	1126	1638	620	1828	4558	2478

–Chitdashwari (1999)

The order of fractions content of sewage biosolid for all the metals as follows :

Zn, Fe, Cd, Pb, Mn: Water soluble < Exchangeable + adsorbed < organically bound < carbonate bound < residual.

Cr, Ni : Water soluble < Exchangeable + adsorbed < carbonate bound < organically bound < residual. (Chitdashwari 1999)

Factors influencing mobility and Adsorption of heavy metals in soils. Understanding the process governing the migration and plant availability of trace metals in soils is essential for predicting the environmental impact of spreading metal containing wastes on agricultural land. The adverse effect of heavy metals is inseparably related to the soil's ability to adsorb and retain sub elements.

pH. The pH of the soil solution maintained at neutral to slightly alkaline condition showed low mobility of all heavy metals. To increase the mobility of heavy metals, the pH of the soil solution should be lowered. The solubility of Pb in soil solution was pH dependent, increasing as the pH was adjusted from 6 to 3. At near neutral pH, the activity of Pb^{2+} showed no clear relationship to pH and a small but significant increase resulting from changing organic matter content. In the near neutral pH range, higher Soil Organic Matter (SOM) increases the Dissolved Organic Matter (DOM), thereby promoting the formation of organo Pb complexes and increasing Pb solubility (Sebastien Sauve, 1998). In acid soil, a mean value of 44% of the fractional sum of Cd is associated with the exchangeable form; there by enhance its mobility. There was about 10 times the amount of Zn on exchange sites where the soil pH > 5.6 Cd (23%) is found in exchangeable form, implying that the metal is relatively very mobile.

In general, sorption increases with increasing pH. That is, the lower the pH value the more metal can be found in solution and thus more metal is mobilized. When pH falls to below 5 mobility is enhanced as a result of the increased proton concentration. At pH values above 7, some heavy metals tend to form hydroxy – complexes which will increase the solubility of the metal in question. Adsorption was greater at pH 6.5 than at pH 4.5. The adsorption of cromate did not change as significantly as other metals with pH changes. This is due to its oxyanion character in oxidising condition, and the adsorption mechanisms are very different compared to that of positively charged metals. These results confirm that adsorption for most metal cations increases as pH increases.

Organic matter. Soil Organic Matter is a key for sorbing phase for metals. The dissolution of humic acid at higher pH is responsible for dissolution of Cu and Pb from soil. Organic matter is important for the retention of metals by soil solids, thus decreasing mobility and bioavailability. However because of the complexation of metals by soluble OM, the addition of OM can result in release of metals from solids to the soil solution.The increase in the solubility of the Cu and Pb is related to the dissolution of the HA component of the organic matter. This indicates that solution phase speciation reactions with organic matter dominate the partition of these metals at higher pH. Because under high pH, the DOM increases as a result solubility of HA.

Higher solubility of heavy metals in soil solution at alkaline pH was attributed to enhanced formation of organic matter metal complexes after ionization of weak acid groups. Extensive evidence exist that in this pH range most dissolved heavy metals are present as metal soluble organic ligand complexes. The degree of stabilization achieved by compost appears to be very important in determining potential mobility of heavy metals. Carillo *et al.,* (2000) reported that, Cd can move readily in sandy loam soils, but OM in the surface layer will act as a sink for Cd and will reduce the rate of movement. Distruction of heavy metals between soil and solute is the key for evaluating the environment impact of the metals. For Cu, in particular OM both solid and dissolved can affect its mobility substantially. (Temming hoff *et al*., 1997) The concentration of DOM is partly controlled by Ca by sorption. Ca can act as a bridge between negatively charged DOM and negatively charged soil particles both clay mineral and oxides.The heterogenity of sorption sites is larges for natural solid OM than for dissolved purified HA.

Naidu and Harter (1998) reported that, the role of organic acids in Cd mobilization is especially important since modern agriculture increasingly involves minimum tillage practices that recycle plant residues. Complexation of metals by organic ligands play an important role in controlling metal solubility. Metal – ligand complex stability generally decreases with a reduction in pH, reflecting the role of $R\ COO^-$ in metal complexation. The ligands include low molecular weight organic acids including oxalic, citric, formic, acetic, malic, succinic, malonic, maleic, lactic, aconitic, and fumeric acids have been identified in soil system. Highest amount of Cd was released by malate and the least by acetate. To separate the pH effects from ligand ion effect, it was found that at high pH values, Cd-ligand ion complexation was essential for the solubilization of Cd.

Ionic strength. The amount of metals adsorbed decreases with increasing ionic strength for minerals with permanent surface charge density (Boekhold *et al*., 1993). Casagrande, (2001) reported that regardless of pH, Zn adsorption was always superior for the most diluted $CaCl_2$, solution utilized. This shows that the reaction took place on the negative surface of colloids. For these surfaces, the reduction of ionic strength makes the surface electric potential more negative, and thus ion adsorption is greater.

In high electrolyte conc. it is also necessary to take into account the competition of Zn and the Ca present in the soil. The more negative electro potential of the surface soil layer explains the greater Zn adsorption at the surface for all pH and ionic strength values, as the surface soil layer presented more negative charges than the subsurface for a same pH values. It is finally concluded that, there was a pronounced increase of Zn adsorption with pH elevation and a reduction of ionic strength of background electrolyte. Results showed that the metal ion was more strongly adsorbed at lower ionic strengths while any increases in electrolyte concentration produced a decrease in metral adsorption.

Adsorption was in relation with the CEC, which obviously could be expected to influence the process at each value of ionic strength. In addition to that, increasing ionic strengths decrease metal sorption because of the competion by other cations for adsorption sites. This soil with higher ionic strength may have more risk of metal leaching from sewage sludge disposal that soil with lower ionic strength.

Soil texture. The soil texture plays an important role in mobility of metals in soil. Texture reflects the particle size distribution of the soil and thus the content of fine particles like oxides and clay. These compounds are important adsorption media for heavy metals in soils. The clay soil retains high amount of metals when compared to sandy soil. Jeanne and Roy (1991) concluded that the fine textured soils contain higher amounts of Pb (3889 mg kg^{-1}) and coarse textured soil contains (530 mg kg^{-1}) lower amount of Pb. Olaniya et al. (1992) reported that the metal content of top and middle layer of soil was comparatively higher than bottom layer of soil. Jeyabaskaran and Sree Ramulu, (1996) concluded that the content of DTPA- extractable metals in light textured soils were low in soils irrigated with sewage water for the past 50 years. The DTPA extractable metal contents of clayey soils (40 years of sewage irrigation) were higher than that of silty clay loam soil (50 years of irrigation)

Pore structure. Alesi et al. (1980) reported that the soil texture, and structure as evaluated by bulk density i.e. the solution flux.This plays a part in diffusion of metal ions into and out of soil aggregates in such a way that movement of elements through soil may be predicated with greater accuracy by having some knowledge of flow velocities. They also stated that flux was least effective for Ni. Facchinelli et al. (1999) found that the downward migration rate of Ni was 0.83± 0.72 cm $year^{-1}$ and was well correlated with high soil macro porosity. Karathanasis (1999) reported that colloids with high surface charge and soils with greater macro porosity contributed more Pb mobility and transport.

Temperature. Several chemical reactions are temperature dependant in the way that they proceed at a metal concentration in plants grown on sewage sludge amended soil. When the temperature raises the metal activity in the soil solution as well as that the plant roots may be more active and has faster absorption rates. Further more, the absorption rate of the roots may be increased as a result of higher evapo transpiration from the plant. Yu Ling et al., (1998) concluded that considerable enrichment of heavy metals in the sub micron range occurs during evaporization of heavy metals in the high temperature environment and the subsequent condensation in lower temperature regions of the systems.

Thermal treatment of Pb^{2+} doped alumina powder could be effective in reducing the leaching level of Pb from the alumina matrix by H_2SO_4 – HNO_3 in deinonized water. The results of the sequential extraction of the thermally treated Pb^{++} doped alumina powder showed that as the treatment duration and temperature increased the percentage of Pb that was bound to the carbonates decreased with a concomitnant increase of residual Pb. Sheaffer etal., (1980) reported that, higher soil temperature enhances the degradation soil organic matter, thereby releasing organic acids, which increase the mobility of heavy metals in soils. Thereby the availability is increased.

Residual Time. Time after application of the sludge, the so-called residual time, may affect the mobility and thus the metal content in the plants growing on it. When the organic matter applied with the sludge is decomposed sorption sites are lost and the accompanying pH decrease also affects the desorption of heavy metals. It has been shown that the metal content increased in successively harvested plants grown on a sewage sludge amended soil. Thus, the available heavy metals in the amended soil may change over time. It has also been argued that a fraction of the organic matter is resistant to degradation and will provide protection against plant uptake and leachability of the metals on a long term basis.

Index cations and anions. The anion which can contribute in reducing heavy metal adsorption by the possible formation of negatively charged or neutral species like: MCl_2, Mcl^-_3, and Mcl^-_4. In particular Cd is known to form relatively stable Cl^- complexes, which greatly influences the mobility of metals in soils. This fact is of particular importance since the ability of Cl^- ions to maintain relatively high concentration of heavy metals in soil solution may produce favourable condition for a faster leaching of metals in the soil profile according to the soil texture or irrigation condition. Avudainayagam et al., (1997) concluded that, the amount of Cr desorbed varied with both the cationic charge and with depth of soil sample. In the surface soils, the desorbed Cr decreased with increasing cationic charge (Water > Na^+ > Ca^{++}). While the reverse trend (Ca^{++} > Na^+ > Water) was observed in the subsurface soils.There was a strong effect of P (Na^+ + 0.003 MP > Ca^{++} + 0.003 MP) in surface soils and Ca^{++} +0.003M >Na^+ + 0.003M> water in sub surface soils. Highest Cr was released in the presence of phosphate solutions and least when Cl^- solution was used as the leaching electrolyte.

On the other hand, the lessened adsorption capacity of soils with respect to heavy metals due to the high concentration of salts could increase the availability of these contaminants to plants. Yujun Yin *et al.* (1995) found that Cl^- ion could be an important factor enhancing Pb and Ni mobility in soil. Mobilization of Pb in contaminated roadside soils is suspected because chlorides are known to complex and solubilizing many heavy metals like Pb and Ni. (Amrhein and Strong, 1990). Approximately 100,000 tonnes of road salt NaCl is applied annually as a deicing salt in the Detroit Metropolitan area (Annon, 1992). Hence it is possible that the observed mobilization is partly the result of Pb complexation with Cl^-.

Smolders *et al.* (1998) found that Pb may also form complexes more readily with NO^-_3 rather than Cl^-. This might be due to the anionic competition between Pb hydroxide species and Cl^- or NO^-_3. Jeffery and James (1993) reported that Pb had been mobilized in vadose zone at the contaminated urban sites studied, and was hypothesized that chlorides derived from deicing salts were responsible. Diyab *et al.* (2000) reported that phosphates and alumino phosphate minerals containing Pb are weakly soluble and quite stable in numerous soils thereby low mobility.

Khan *et al.* (2000) indicated the presence of NH_4^+, Na^+, K^+ and Ca^{++} as cations and NO_3^-, Cl^-, SO_4^{2-} and PO_4^{3-} as anions released from applied fertilizers could be regarded to play a dominant role in mobility of Pb and Ni in soil because of blocking of adsorptive site by the above said ions. Faud Ababneb *et al.*, (2001) reported that the major cations Ca^{++}, Mg^{++}, Na^+, K^+ at the high concentrations that they are present in the water samples have greater effect in releasing Ni^{2+} ions from DOC bound Ni thereby increased mobility. Smolders *et al.*, (1998) reported that, neither osmotic stress at high salt levels nor ion exchange of Na^+ or Cd^{2+} are mechanisms that explain the effect of NaCl on increasing Cd concentration in plants. Mobility of Cd in soil is increased at higher Cl^- concentration due to the increased concentration of Cd in the liquid phase. This effect can be important if the transport of Cd in soil toward plant roots limits the uptake from soil. The effect of Cl^- on Cd uptake could be explained by (*i*) $Cd\,Cl_n^{2-n}$ species which do not adsorb on soil or clay in solution are also available for plant uptake (in addition to Cd^{2+}). (*ii*) Cl^- enhances diffusion of Cd^{++} through the apoplast to sites of Cd uptake to the root itself.

Amendments and Additives. Friesl *et al.*, (2000) found that in unpolluted soils, the mobile fraction of heavy metals is small compared to total concentration however may increase considerably in polluted soils and hence be hazardous to the ground water or food chain. Due to the lack of low cost, environmentally sound decontamination technique for heavy metal contaminated soils the immobilization by soil additives can provide a technique to lower the risk of metal mobility, bioavailability and toxicity. The Shale can be used as an amendment in soil detoxification because it possesses great sorbability, and markedly reduced the mobility of heavy metals.

Table 3 : Effect of amendments on NH_4 NO_3 extractable and plant uptake of metals in soils (mg kg^{-1}).

Soils	Test	Zn		Cd		Ni	
I	**Control**	1.63	b	0.11	b	0.65	b
	+ red mud (1%)	0.60	a	0.06	a	0.32	a
	+ Zeolite (2%)	1.54	b	0.11	b	0.62	b
	+ lime (0.3%)	1.41	b	0.10	b	0.61	b
II	**Control**						
	+ red mud (1%)	159.1		4.03		2.56	
	+ Zeolite (2%)	149.6		3.77		2.67	
	+ lime (0.3%)	158.9		4.22		2.96	

Additives used are, red mud (10g kg^{-1} soil), Zeolite (20g kg^{-1}) lime (3g kg^{-1}). Red mud was the only amendment that decreased extractability and pH uptake of Zn, Cd and Ni continuously. Red mud performed well in the 15 months pot study but the results have to be evaluated in long term field experiment. EDTA enhanced phyto remediation assisted by strong, barely degradable chelates is likely to increase the risk of ground water pollution during the phyto extraction process.

Table 4 : Effect of gypsum on the transport of trace metals (mg kg^{-1}).

Treatments	Cu	Zn	Cu	Mg	DOC
Control	0.32	0.12	272.0	46.1	83.2
Gypsum (4.g Mg ha^{-1})	0.15	0.06	445.1	70.9	48.7
Gypsum (9 Mg ha^{-1})	0.10	0.08	572.5	92.3	33.4
Mg So_4 (6.3 Mg ha^{-1})	0.23	0.09	509.9	228.5	72.8
K_2So_4 (9.2 Mg ha^{-1})	0.49	0.18	399.6	65.4	140.8

Zhu and Alva, (1993).

Transport of considerable amount of Cu and Zn from the unamended soils was indicated by the recovery of these metals in the leachate fraction. Fig. In sandy soils amended with gypsum or Mg SO_4, Concentration of Cu and Zn in the leachate fractions decreased significantly compared with that from the unamended soil. At equivalent rates of cation addition, the transport of Cu and Zn from the soil amended with gypsum was less than that from the unamended or Mg So_4 amended soil; however application of K_2SO_4 increased the transport of Cu and Zn compared with that from the unamended soil.

In order to investigate the role of organic ligands in transport of Cu and Zn, concentrated of DOC was measured in the leachate. Cumulative DOC recovered the leachate was greater in the K_2SO_4 amended soil than that in other treatment. This implied that the gypsum or Mg SO_4 amendments decreased the transport of DOC. Increased organic carbon in the leachate may have increased the chelation of metals, resulting in an increase in their solubility and transport. Soil management factors that increase dispersion of organic matter and transport of DOC eg. K_2SO_4 amendment in this study, is more likely to increase the transport of trace metals in soils. On the other hand, application of gypsum or Mg SO_4, improves soil physical and chemical properties thus minimizing the transport of DocOC and the metals complexed with it. Myrtveit (1999) reported that, mobility of heavy metal is enhanced by increasing acidity. But liming decreased the mobility of Zn and Cd in organic horizon.

The capacity of several additives *i.e.* Thomas phosphate basic slag, hydrous iron oxide (HFO) , hydrous manganese oxides (HMO) and Beringete were tested to see their immobilizing effect of Cd and Pb in soil (Michel et al., 1994). The maximum decrease in mobility and plant uptake of Cd and Pb should be mainly related to the solubility and sorption mechanisms, particularly the reactive sites on the surface layer of HMO that could bind Cd or Pb. (Khattak and Page, 1992). Point zero charge (PZC) values for HMO ranged from 1.5 to 2.0, whereas those of HFO were 8.0 –

8.5. At pH values, higher than the PZC, HMO is negatively charged and has a CEC that increases with increasing pH. Therefore HMO has a high adsorption capacity for Cd. Also, reduction in Cd uptake can be partly related to the Mn added with HMO, causing increased Mn conc. in plants from HMO treated soils. It is known that Cd and Mn ions compete during the uptake process and that the effect of Cd on plant metabolism could be reversed by Mn application.

Slope. Korentajer *et al*., (1993) reported that, slope affects the amount of sediment yield and run off volume and therefore the amounts of Cd transported from the contaminated soil in the various phase. Transport and fractionation of soil aggregates taking place during the run off event. At low slope angle and low runoff flow velocity soil loss is mainly due to sheet flow as a result of inter rill erosion. Sheet flow lacks sufficient velocity to transport sand particles, which consequently, during sheet flow, clay sized particles are transported in preference to sand particles, resulting in a selective enrichment of the sediment with clay and Cd.

Table 5 : Effect of slope on clay and Cd enrichment ratios in the sediment.

Soil	Slope %							
	10		20		30		40	
	Clay	Cd	Clay	Cd	Clay	Cd	Clay	Cd
Jogine	1.61	1.83	1.92	1.11	0.99	1.19	0.77	0.63
Msinga	1.37	1.43	1.22	1.36	1.24	1.24	1.22	1.19
Nbgagu	1.11	1.05	n.d	1.02	n.d	1.00	1.11	1.00

Korentajer *et al*.,(1993)

The effect of slope on sediment Cd enrichment ratio was, in general, similar to the effect of slope on clay enrichment ratio indicating that Cd is transported mostly in the clay fraction.In highly eroded soil (Mngagi) as well as low erodabiity soil (Msinga), an increase in slope did not significantly affect the clay enrichment ratio indicating that Cd is transported mostly in the clay fraction. In highly eroded soil (Mngagi) as well as low erodabiity soil (Msinga), an increase in slope did not significantly affect the clay enrichment ratio and the concentration of clay associated Cd in the sediment. This finding underscores the danger of surface water contamination by sediment associated Cd transported from areas contaminated by surface deposition of Cd. In particular, process such as replacement of adsorbed Cd by Ca^{++} (for eg. addition of phospho gypsum) and decrease in soil surface pH (due to acid rain) may increase the soluble Cd runoff load.

Metal Speciation. Sadler *et al*., (1994) reported that As mobility in soil is affected by the species of As present and the nature of the soils involved. As (111) is more mobile than As (V).

Deicing Salts. Amrhein *et al*., (1994) reported that, Calcium magnesium Acetate (CMA) is an alternative to NaCl for snow and ice removal on roads and at airports (Trans. Res. Board, 1991). However, there has been some concern that CMA may increase trace metals mobilization from road side soils, because acetate is stronger ligand than Cl^{-} and divalent cations may displace metals from exchange sites. Amrhein and Strong (1990) showed that the aerobic decomposition of the acetate in CMA resulted in the precipitation of CaCO$_3$ and the immobilization of Cd as a Cd precipitate with the Ca CO$_3$. Low redox conditions resulting from CMA decomposition could have a strong influence on trace metal solubility as well.

Soil Colloids. Karathanasis (2000) reported that, most heavy metals are generally considered relatively immobile in sub surface soil environment. However, under certain condition their mobility may be accelerated by potential contributions by mobile colloidal particles acting as metal carriers or facilitators to ground water. Contrary to initial findings emphasizing organic colloids as the primary contaminant carries; more recent research has documented similar or greater potential for metal facilitated transport by mineral colloid particles. The presence of colloids increased the transport of both soluble and colloid sorbed metal fraction. Colloid mediated transport increased with colloid surface charge, pH, organic carbon and soil macroporosity and was inhibited by increasing colloid size., quartz and sesquioxide content. Metal mobility and load carrying capacity followed the mineralogical sequence montmorillinite > mixed > illitic > Kaiolinite.

Flushing Pb–contaminated soil columns with uncontaminated colloid suspensions resulted in Pb–desorption and remobilization up to 50 times greater compared to that of de ionized water solution. Organically enriched colloids were less effective metal desorber than mineral colloids due to their lower binding energy, but caused greater soluble Pb mobilization through formation of organo metallic complexes. A stronger specific metal sorption affinity for the colloid surface than for the soil matrix appeared to be the dominant mechanisms facilitating metal transport.

Competing ions. Competing ions can have a marked effect on ion sorption by soils. Chien *et al*. (1992) found that presence of Pb did significantly reduce the adsorption maximum of Cd on soils. Fernandez *et al*. (1999) found that there is a positive correlation exist between Mn and Ni, Ni and Cu, Ni and Fe, Pb and Cu, Pb and Fe, Pb and Mn in soils.The presence and competitiveness of Cr (III) in a multiple ion mix will greatly reduce the amount of Pb^{2+} sorbed, thereby drastically increasing their mobility and potential for transport to ground water. Mandal Rupasri (2000) reported that, the stability constant for the Pb^{2+}- FA complexes decrease with increasing concentrations of trace metals such as Cu^{++} and Mn^{2+} and Ni^{2+} – FA by Ca^{++} and Mg^{++}. This might be due to the polyelectrolyte effect. Minkina and Samokhin (2000) found that a decrease in Pb adsorption is related to the abundance of Ca^{2+} ions as well as other cations available in the solution.

Harter (1992) reported that Ni^{2+} and Co^{++} compete equally with Ca^{++} only at very low solution activities. The Ni^{2+} adsorption was accompanied by an equal Ca^{++} desorption indicating that Ni^{2+} ions were retained at sites previously occupied by Ca^{++}. The Ni^{++} sorption has no effect on Cu^{++} adsorption by reducing a Mn ion, creating an adsorption site. The Ni^{2+} should compete with Co^{2+} (but not Co^{3+}) for site occupation and Co^{2+} sorption should be depressed by increasing Ni^{2+} activity.

Synthetic chelators. D'Souza et al (1983) reported that leaching the soil with dilute solution of 10M EDTA, EDDHA and DTPA resulted in enhancement of the mobility of all these heavy metals with a high degree of chelate specificity for individual ions depending upon the soil type. Rapid formation of stable soluble Cr-EDDHA, Pu-DTPA and AM – DTPA complexes facilitate the leaching of these pollutants from the contaminated soils.

Microbial metabolites. Wildung *et al.,* (1979) reported that, soil bacteria and fungi produced soluble exocellular metabolites which complexed Ni on elution of exocellular solutions through soil column. The mobility of complexed Ni on elution of exocellular solutions through soil was depend out upon the charge and molecular weight of the complex, the affinity of the ligend for Ni the conc. of Ni in solution. Neutral and anionic Ni complexes with apparent molecular weight less than 1200 arising from fungal metabolism were most mobile in soil increasing Ni mobility by up to 1000 times relative to inorganic Ni. A single Ni complex responsible for the most pronounced increases in Ni mobility in soil consisted of several ligands capable of complexing Ni independently and exhibiting both positive and negative charge after separation from the original complex ligend.

Phenolic substances. The mobility of heavy metals was largely influenced by the chemical structure of the phenolic acids. The affinity of pthalic acid to metals was much less than that of phenolic acids.The amount of mobility with salicylic and pthalic acids increased as the soil pH decreased. The results showed that chelating phenolics such as proto catechuic acid and salicylic acids, which were exuded from plant residues or produced during the decaying process of plant residues enhances mobility of Fe, Al, and Mn from soil particles and accelerated the downward movement of these metal ions.

Herbicides. Morrillo *et al.* (1999) reported that Glyphosate has three groups (amine, carboxylate, phosphonate) that can form strong coordination bonds with metal ions, particularly with the hard transition metal ions. This ability places glyphosate in an almost unique class of strongly chelating herbicides.

The addition of urban waste compost to the soil increased the Cu adsorption capacity. The cu retained by the soils cannot be removed passing an electrolyte solution through the soil column, but it is removed to a great extent by passing a glyphosate solution. This is probably due to the formation of very strong Cu-glyphosate complexes in solution that have lower tendency to be adsorbed on the soils than free cu. It indicates the high extracting power of this herbicide showing its potential in remediation of metal contaminated soils.

Fertilizer and soil amendments. Raven and Laeppert (1998) reported the trace element concentration of different fertilizers and amendments. Heavy metal concentrated N fertilizers including urea, ammonium nitrate and calcium nitrate have been reported to range between 0 and 3000 μg g^{-1} (Popova, 1991; Severin *et al.* 1990).

Table 6 : Heavy Metal composition of fertilizer and soil amendments.

S.No.	Sample	Cdμg g^{-1}	Coμg g^{-1}	Crμg g^{-1}	Niμg g^{-1}	Pbμg g^{-1}
1.	Urea	<0.2	<0.7	ND	<0.2	<0.4
2.	Ammonium nitrate	<0.2	<0.07	ND	<0.2	<0.4
3.	Ammonium sulfate	<0.3	<0.07	2.14	<0.2	<0.4
4.	MAP	<0.3	0.78 ± 0.62	16.9	0.6 ± 0.4	<0.2
5.	DAP	4.6	3.24 ± 0.30	ND	22.2	2.9
6.	Triple Super PO$_4$	5.0	6.61 ± 0.49	88.9 ± 4.8	25.2	11.1
7.	Kcl	<0.2	< 0.07	< 1.05	<0.2	<0.4
8.	North Corolina	RP 48.8	< 0.08	140 ± 7	16.8	4.6
9.	Tilemsi RP	1.3	19.6	33.2 ± 3.2	50.4	29.2
10.	Calcite	0.7	< 0.08	ND	1.4	1.1
11.	Dolomite	<0.2	0.03 ± 0.09	32.3 ± 1.8	3.3	0.7
12.	Manures	0.7	2.23 ± 0.54	ND	ND	0.7
13.	Compost 1	0.5	3.57 ± 0.69	14.4 ± 1.3	14.4	7.5

–Raven and Laeppert (1997)

A similar trace element concentration range has been reported for K fertilizer such as potassium chloride and sulfate and for composted municipal waste (Sposito, 1983; Popova, 1991). The trace element concentration in manure samples have been reported to reach 900 µg g^{-1} (Popova, 1991). Trace metal concentration upto 10,000 µg g^{-1} have been reported for ammonium PO_4, Super PO_4, liming material and sewage sludge samples (Amaral Sobinho et al., 1992). RP generally had the highest reported trace element concentrated leaching the 100,000 µg g^{-1} level. (Amaral Sobrinho *et al*., 1992).

Adsorption and Retention of heavy metals in soils. The adsorption maxima is positively correlated with pH and organic carbon content. This is because concentration of monovalent Cu oH^+ ions which are known to be adsorbed in much greater quantities than divalent (Cu^{++}) ions which increases rapidly with increasing pH of the medium. Besides, increase in pH decreases positive charge and or increase negative charges on the soil colloidal surface. This is more so in case of Alfisols which are known to have high amount of pH dependent charge. The positive correlation between adsorption maxima and OM content of the soil might be explained by the fact that Cu could get complexed very strongly with the latter because of its unique ability to form rigid inner sphere complex.

Langmuir adsorptiopn equation

$$C/x/m = 1/Kb + C/b$$

Freundlich adsorption equation

$$\log (x/m) = \log kf + 1/n \log C$$

Where in C = Equilibrium heavy metal concentration (ìg ml^{-1}) ; x/m =Amount of metal sorbed by soil (šg g^{-1}) ; b = Sorption Maxima ; K = Bonding Energy (L mg^{-1})

Freundlich Adsorption Isotherm. The Freundlich Kd values which are analogues to stability constants for complexes, correlated significant and negatively with pH (r = 0.84^{xx}) and organic carbon. With increase in pH, Kd increased due to increased ionisation of functional groups on organic acids and of binding sites on oxide surfaces. (Saha et al., 1995).

Simultaneous sorption of Cd, Cu, Ni, Pb and Cr on soils treated with sewage sludge *supernatant*. Components that partition more strongly to the solid phase exhibit low mobility in soils while those that partition more strongtly to liquid phase have greater mobility and may ultimately contaminate the ground water. Accumulation of heavy metals in surface soils has been reported from SS application.

Complexatn of metals by organic ligands is one of the mechanical controlling the available and mobility of metals. Formation of soluble organs metal complexes increases availability and mobiity of trace metals.

On the other hand, formation of metal complexes with solid phase organics may increases the partitioning of metals toward the solid phase. Therefore OM may be an important factor influencing the retension of trace metals by soils.

Stability of complexes varies among metals and among organic ligands for the same metal. Cu forms a relatively strong 1:1 complex with acidic functional groups in fulvic acid extracted from sewage sludge complexation of Cu by the carboxylate group of HA results in formation of a unidentate complex (*i.e.*, a single M – O bond). Cd complexatn with organic ligands is much weaker than that for Cu. Sorption affinities of metals to solid – phase surfaces are very different among metals which is reflected by the sorption maxima and competition among ions.

Eg. Cu adsorption was not significantly affected by Zn addition upto 250 µg mL^{-1} , although the presence of Cu even at a concentration of 15 µg mL^{-1}, completely prevented Zn sorption in one soil.

Sorptn of Cu, Ni & Co occurred in the order of Cu > Ni ~ Co in a Ca – saturated soil (Harter, 1992). Cu caused a significant decrease in Ni sorption, but Cu sorption was not affected by Ni. It was concluded that Ni was retained by an exchange mechanism, but Co and Cu sorption was more complex (Harter, 1992). Addition of Ni or Cu only affected Zn adsorption when metal concentration in solution were in excess of the soil sorption capacity.

The correlation between SOM and the sorption of the metals may be explained by formation of organo –metal complexes. To estimate the stability of organic complexes with metals of the contributionn of metal partitioning between solid and solution, speciation of metals at pH 4.5 and 6.5 is necessary.

Metal complexes c acetate also increase from pH 4.5 to 6.5 while hydroxyl forms of the metals are negligable. At high pH, the stronger adsorption results from the dissociation of functional group on the organic phase due to less competition from H^+. Thus, the sorption trend of the metals was a result of a combination of metal complexes with both DOM in the solution phase and organic matter on the solid phase, as well as competition among metal ions. In the case of disposal of sewage sludge, high OM concentration are introduced to the soil system. The DOM could be adsorbed rapidly on the soil.

Cr was the most weakly adsorbed ion among all the elements and there was no correlation between Cr sorption and SOM content at either pH 4.5 or 6.5. This is due to hexavalent form (CrO_4^{2-} or $Hcro^{-}4$). The CrO_4^{2-}fraction increase and $HCrO_4^{-}$ decreases as pH changes from 4.5 to 6.5. It's adsorption should decrease as pH increaess due to increase of negative charge on the solid phase. In contrast to theory, sorption of Cr was higher at pH 6.5 than at pH 4.5; Pb was most strongly adsorbed element. Studies of Pb chemical fraction and solubility on 3 surface soils containing high Pb levels showed that <0.07% of the Pb was in exchangable form. Therefore Pb was present dominantly in non exchangeable forms associated primarily with the organic and residual fractions in two of the three soils and predominantly specifically bound to inorganic sites in the third soil. The results suggested that 99% of the Pb partitioned in the solid phase. Cd and Ni are weekly adsorbed metals. Cd contamination has the greatest potential for limiting application of sludge on land because of its relatively high mobility in the soil environment. The sorption of Cd was studied in three soils that were unamended, freshly amended, or pre-conditioned with γ – irradiated sewage sludge. Results showed that Cd sorption was greatest in unamended soils, less in soils preconditioned with sludge and the least in the freshly amended soil.

Comparison among Kd values for all the soils and the amount of metal adsorbed at the 50 µg mL^{-1} , addition indicates the following selectivity of metals.

Pb > Cu > Zn > Ni > Cd ~ Cr at pH 4.5

Pb > Cu ~ Zn > Cd > Ni > Cr at pH 6.5

Pb, Cu and Zn – strongly adsorbed

Cd, Ni & CrO_4 – weakly adsorbed

Therefore leaching of Cd, Ni, CrO_4 is most probable for this soils studied. Disposal of sewage sludge in this study is unlikely to increase leaching of Pb, Cu, Zn in these soils in the short term, especially due to the near neutral to alkaline pH values.

- The risk of elevating certain element levels such as Cr, Cd and Ni still exists depending on the sorption capacity of the soil, the composition of sewage sludge and application rate and frequency.
- The metal sorption strength at low metal loading was related to OM concentration and surface area. This suggests that SOM plays an important role as well as pH.
- Further disposal of sewage sludge has been reported to raise SOM concentration which may alleviate the risk of metal contamination to a certain extent.
- However long term effects of sewage sludge disposal on the environment need further studies under specific environment condition.

Table 7 : Distribution coefficient (Kd) for Cd with increasing Ca Concentration

Ca Concentration M/L	Distribution	Coefficient (Kd)	
	Lamporecchio	Sobretta	Prentano
0.005	0.14	1.97	0.92
0.010	0.06	0.75	0.70
0.025	0.05	0.55	0.55
0.050	0.02	0.35	0.44

Source: (Suduan gao et al., 1995)

At low concentration of Cu had such a high affinity for the adsorbing surfaces of the soil that it is completely adsorbed. As the concentration of the metal increases more sites on adsorbing surfaces of soil are filled it becomes more difficult for Cu to find available sites. In these soils, the quantity of adsorbed metal approaching the saturation at every value of ionic strength. This is observed in the case of Orentano and Sobretta soils (Freundlich type).

In the case of Lamporecchio soil adsprption isotherm could be classified as Langmuir type showing a lower affinity of this soil for the metal. Also, in this soil adsorbed Cu seems to reach saturation which means that the metal had filled all possible available sites and further adsorption could take place only on few surfaces.

To conclude, the decrease of Cu and Cd adsorption with increasing ionic strength can be attributed mainly to the increasing competition of Ca for the adsorption sites of the soil. Moreover, the adsorption of Ca may lead to an increase of positive charges at the soil adsorbing surfaces and this in turn reduces the heavy metals adsorption.

ADVERSE EFFECTS OF METAL MOBILITY

Heavy metals in ground water of Madras City. *Selvapath and Lakshmi, (1995) reported that out of the 76 water samples Cd was present in two sample but the levels are above the acceptable limit. Though Zn was present in majority of the samples, the percentage of samples in which the levels were above the acceptable limit never exceeded for Fe and Mn were not only detected in majority of samples but their levels were also above the acceptable limit in most instances. Cu was detected in significant number of samples but only in few samples the levels exceeded the acceptable limit. Ni was detected in a sizable number of samples and in > 88% of them the levels were above the guideline value of 0.02 mg/l.*

Impact of textile industries on the ground water quality of Tirupur.

Contamination of Noyyal river. Thompson et al., (1995) reported that, Noyal river a Tributory of Cauvery, Dye mid – effluent samples contain, higher amount of pb. The studies on the ground water quality of industrial site of Tiruppur, suggested that effluents are let into the dry noyyal river the environmental issues are prominent. The concentration of heavy metals like Zn, Pb, Cd were also higher when compared with the Indian standards for drinking water. The heavy metals like Zn, Pb, Cd exceeded the permissible level in both tube and open wells. Ground water mining association the industrial is common in this area. Non point sources, especially run-off from these index units are how by far the principle sources of both environmental and toxic water pollutants in this area.

Mobility of heavy metals and ground water quality. (Patel and Shrivastava, 1999) found that, the textile, dying and printing industrial effluents have an adverse effect on the soil ground water eco systems. It is due to leachates of cationic and anionic species from textiles effluents through soil strata. A comparison of collected data clearly indicates that about 3-8 fold increase in the conc. of all parameters of effluent amended soil that control site.

Textile, dying and printing industrial effluent:

Ca > Na > Mg > h > Zn > Cr > Fe > Hg > Pb > Cu > Cd > As > Ni

Ground water near industrial area

Ca > Mg > Na > k > Zn > Fe > Cr > Pb > Hg > Ni > Cu > Cd > As

Soil affected by industrial effluents

Ca > Mg > Na > k > Fe > Zn > Cr > Cu > Pb > Ni > Hg > Cd

Unless, this soils are managed properly, it may lead to severe land and ground water pollution affecting various componentt of the soil eco system.

Cr Contamination of water resources and soils of Carpet industrial area of eastern Uttar Pradesh. Singh et al., (2001) reported that, high levels of Cr in ground water samples indicate the leaching of metal released from the industry. It has been shown that Cr (V1) has high mobility than Cr (III) and that the latter is preferentially retained by the soil. They have also shown that a part of Cr (III) can be oxidized in the prevailing soil conditions to Cr (VI). The waste water of carpet industry contains both these forms and thus pollution of soil by adsorption of Cr (III) and that of water by Cr (VI) becomes possible from one polluting source.

Generally, contaminate move in soil by preferential glow *i.e.*, this flow can enhance chemical mobility by moving contaminants through macropores instead of moving them through soil matrix where they could be adsorbed, chemically degraded or biologically transformed. There is a positive correlation between heavy metal in soil and coarse sand content.

The downward migration of Cd, Pb and Cr were positively correlated with fine sand content, suggesting the mobility of trace metals is enhanced by high soil macro porosity.

Studies on the concentration of Pb and Cd in milk of Cow and Buffalo in Madras City. (Ayyadurai et al., 2000) concluded that,

1. Buffalo milk contained higher levels of Pb in comparison to that in cow milk in all the samples analysed.
2. Pb concentration in Parrys and Anna Nagar area were higher than those in Adayar, Mambalam, triplicane.
3. Pb concentration in milk showed appreciable difference between zones.
4. The level of Pb in all the samples were below the stipulated permissible limit.
5. Cd concentration was below detectable limit in all samples investigated.

Table 8 : Lead concentration in the milk (µg/l) of cow and buffalo in difference zone of Madras city.

Zones	Cow milk	Buffalo milk	Mean for zone
Adyar	6.98 ± 0.91	7.53 ± 0.87	7.26 ± 0.63
Mambalam	2.28 ± 0.75	8.98 ± 1.16	5.63 ± 1.02
Triplicane	6.46 ±.1.23	7.17 ± 1.43	6.82 ± 0.94
Parrys	8.74 ± 0.62	16.27 ± 2.19	12.11 ± 1.74
Anna Ngr	8.49 ± 1.74	13.38 ± 1.93	10.94 ± 1.41

- Buffalo milk contailned higher levels of Pb in comparison to that in cow milk in all samples analysed.
- Pb concentration in Parys & Anna Nagar were higher than those in Adyar, Mambalam, Triplicane.
- Pb concentration in milk showed appreciable difference between different zones.
- The level of Pb in all samples were below the stipulated permissible limit.
- Cd concentration was below the detectable limit in all samples investigated.

Conclusion. In seeing all those things, mobility of heavy metals is mainly influenced by the soil factors such as pH, organic matter, ionic strength, CEC, texture, temperature, salinity amendments, Zn fertilization, rhizosphere pH , anionic spp, ligands, phenolic substances, herbicides, organic pollutants , microbes and so on. Mobility of metals is controlled by sorption process. The adverse effects of mobility are , they contaminate ground water and ultimately causes human health hazards. Their persistent in soil also causes accumulation of heavy metals in food chain when they reach the conducive environment which is suitable for conversion of unavailable form into available form. Hyper accumulator plants which may extract excess amount of metals from soil ie. phyto remediation. Identification of plants which can accumulate more quantity of heavy metals in their tissues are the prime most important technique to clean the contaminated soil environment.

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