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Study of Interaction of Loamy Soil with Salts of Heavy Metals in Assessment of Groundwater Contamination

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ABSTRACT

A major environmental problem is the risk of metal pollution of surface and groundwater due to leaching from polluted land-based solid wastes. The goal of this research is to figure out what happens when heavy metals interact with sandy and silty loam soils, and how that affects groundwater quality. As the sorption decreased in the order Mn> Fe> Ni > Cr> Zn> Cu> Pb> Cd. The leaching order should be reverse of it. As a result, Cd is less well held by soils than the other hazardous cations, and hence poses a greater risk of polluting groundwater due to its severe toxicity. The metals Zn, Cu, Pb, and Cd were shown to be significantly more mobile than the other four. The retention of metals became weaker as the concentration of metals increased. This points to the probability of groundwater contamination as a result of land infill. As a result, at all polluted sites in Agra, they should be properly monitored. Metals are more sorbed on silty loam than on sandy loans because the former has higher organic matter, clay, monmorillonite contents, CEC and surface area. Because the majority of the soil in Agra is sandy loam, it is recommended that all wastes containing heavy metals be treated before being disposed of in loamy formations.

Keywords- Environment, Groundwater, Loamy Soil, Heavy Metals.

I. INTRODUCTION

In general, municipal solid waste landfills pollute the environment significantly due to landfill gas combustion, leachate, leakage, and bad odours. Because leachate contains large concentrations of heavy metals, organic compounds, and hazardous substances, it has the greatest impact on the surrounding ecosystem, particularly surface and ground water bodies. Several examples of pollution of aquatic bodies caused by municipal solid waste landfills have recently been documented around the world.[1]

Heavy metals have risen to prominence in recent years as a result of their non-biodegradable, dangerous, and toxic characteristics. Almost 70% of industries use heavy metal compounds in their day-to-day operations. Electroplating, for example, releases a large amount of metals into the atmosphere. Leaded gasoline, which has concentrations of 1000-4000 mg/kg on busy streets, enriches roadside soil with Pb. Since ZnO and zinc dialkylcarbamates are used in the vulcanization process, rubber tyres may contain 0.09 mg/g of Cd, and Cd occurs naturally with Zn. The abrasion of tyres on the road adds Cd to street dust, which is deposited in soil by both wet and dry processes. Despite the fact that Cd mobility and plant availability in soils are limited, its severe toxicity can be a serious problem. Batteries contain lead, cadmium, and mercury. Metal plating uses Cd, Cr, Cu, and Zn. Metals such as Fe, Cu, and Zn are commonly used in wire, rod, brass, bronze, and medicine. Hg is utilised in the chloralkali industry, electrical equipment, and seed dressing fungicides. Hg(0), Hg(1), Hg(II), and Cd(II) are less strongly held by soil than the other hazardous cations, posing a greater environmental threat.

In India, about 3 lakh tonnes of urban solid waste (USW) is generated everyday from major urban centres. Hyderabad with a total population of about 50 lakh produces about 2000 metric tons of USW daily [2]. About 600 metric tons of non-segregated garbage is dumped in Shahadra landfill and 100 tons in Lohamandi. Shahganj and Bundu Katra landfills daily in Agra [3] having about 22 lakh population. USW can cause groundwater pollution [4]. Groundwater of Agra city is not fit for drinking because it has hardness, Ca, NO_3 and F values exceeding the limits set by national and international agencies. [5] The landfill pollution show leaching of various organic and inorganic chemicals into the groundwater. [6]

A study done by Zhou, Y., Li, P., Chen, M. *et al* assessed the carcinogenic and non carcinogenic health effects of groundwater pollutants in Gu'an County, North China Plain area. They found that for adults, only 12.5% of the 16 shallow groundwater samples are above the acceptable potential non-carcinogenic risk level, while 87% of the samples exceed the allowable level for children in shallow groundwater. The non-carcinogenic risk is mainly posed by F^-, Fe and As. Regarding deep groundwater, no non-carcinogenic risk is observed for adults, but Cr^{6+} contributes to non-negligible non-carcinogenic risk for children. [7]

Groundwater is a valuable resource for drinking and irrigation, as well as a major source of human contamination. A study conducted by Narshima et. al. in the hard rocky terrain of Southern India on 43 groundwater samples, found that the groundwater nitrate and fluoride contents vary from 2.2 to 165 mg/L and

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0.84 to 0.85mg/L, respectively. The non-carcinogenic risk ranges from 0.98 to 8.29 for infants, 0.85 to 7.18 for children, and 0.63 to 5.31 for adults. Infants and children are more vulnerable than adults in the study region [8]. The nitrate pollution is a serious threat to infants, causing a disease known as blue baby syndrome which affects the ability of blood to carry oxygen and may lead to death [9]. The data, which are obtained from Department of Fertilizer, Govt. of India, shows that the use of nitrate fertilizer has been increased in last 16 years. Although the fertilizer loss is important economically to the farmer, the more important concern is its effect on the environment.

Sudarshan kurwadkar et. al. documented groundwater quality impact during the year 2019 due to both natural and anthropogenic activities throughout the world. In this article inorganic, organic and microbial pollutants were discussed and also remediation techniques for inorganic pollutant were reviewed. [10]

Groundwater quality of Agra district has been assessed by Vinod kumar et.al. (2017) considering twelve water quality parameters. Data on groundwater quality of fifteen blocks were collected for nine years (2006-2014). The data were investigated using Wilcox and Piper diagrams with the help of Aquachem 2011.1 software. The assessment on suitability of groundwater for irrigation purpose was done using sodium percentage (Na%), Sodium Absorption Ratio (SAR) and Residual Sodium Carbonate (RSC) for all the blocks. The results show that groundwater of various blocks of Agra district contain the salts of sodium, potassium, calcium and magnesium[11]

High values of Cd, Pb and Zn in industrial areas are probably due to galvanising, electroplating, foundry, beverage and other industries and in dumping areas due to leaching of these metals from dumping sites.

In comparison to air and water, soil is regarded a safer medium for waste disposal since it can absorb and oxidise contaminants to their least dangerous forms [12]. Except for NO_3^- and CrO_4^{2-}, ion leaching is generally regarded minimal. The Ganga and the Yamuna basins have multiple and shallow aquifers that have been reported to be contaminated. The unsaturated zone and shallow aquifers are mostly of silty loam soil but sandy loam dominates in the Agra region of the Yamuna doab bordering Rajasthan. Silty loam sorbs more than sandy loam due to higher clay, $CaCO_2$, OM, CEC and surface area. So high leaching is expected in Agra region and revealed by abandoned wells, tube wells, jet pumps and hand pumps. The aim of this study is to know what happens when Cr. Mn, Fe, Ni, Cu, Zn, Cd and Pb interact with sandy loam and silty loam soils and how they affect groundwater quality.

II. MATERIALS AND METHODS

A composite sandy loam sample was taken from St. John's College's residential garden trench-fill, as well as silty loam soil from the Shahadra landfill. Soil samples were taken with a screw auger in zig-zag along different transacts from the middle horizon (0.15-0.30 m) and the deep horizon (1.8-2 m) [13]. By diagonal quartering, a 2 kg composite sample was obtained from 10-20 sub-samples in a polyethylene bag with a 4-kg capacity. Standard method [14] were used to establish their attributes, as shown in Table 1.

Table 1: General properties and heavy metal contents of the loamy soil

Soil Characteristics	Sandy Loam		Silty Loam	
	Middle Horizon	Deep Horizon	Middle Horizon	Deep Horizon
Physical				
Texture	Sandy Loam	Sandy Loam	Silty Loam	Silty Loam
Sand%(2000-20μ)	60.0	65.0	48.0	50.0
Silt % (20-2μ)	24.0	23.67	35.20	32.44
Clay %(< 2μ)	16.0	11.33	16.80	17.56
Physico-chemical				
pH (sand :H2O=1:5w/v)	7.4	7.90	7.50	7.70
EC(dS/m (sand:H_2O=1:5w/v)	3.80	0.34	3.75	0.58
$CaCO_3$ (g/kg)	10.30	13.30	18.40	15.26
Total soluble salt (%)	0.09	0.07	0.10	0.09
Organic Carbon(%)	11.9	6.78	15.8	18.79
CEC (meq/100gm)	8.98	12.90	8.00	26.38
Surface area (m^2/g)	76.20	67.35	216.10	199.26
Surface charge density (meq/g)	13.26	11.23	16.26	13.32
Total Metal Concentration				
Cr	318.7	22.3	293.0	175.0
Mn	422.2	168.88	836.0	167.2
Fe	86320.0	47476.0	89000.0	3560.0
Ni	136.0	85.0	251.0	125.5
Cu	498.0	441.97	498.7	423.89

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Zn	522.0	443.7	595.0	476.0
Pb	398.0	368.0	320.0	288.0
Cd	3.80	3.65	5.20	5.2

All chemicals used were analytical grade. Throughout the experiment, distilled deionized water (DDW) was used. For instrument calibration and sorption studies, four standard solutions of 1, 10, 100, and 500 mg/l concentrations of Cr, Mn, Fe, Ni, Cu, Zn, Cd, and Pb were prepared by diluting their stock solution of 1 g/l, i.e. 1ml=1 mg metal. The stock solution of Mn, Ni, Fe, Cu, Zn and Cd were prepared by dissolving 1.000 g of 99.5% AR 325 mesh metal powder from CDH, New Delhi in a minimum volume of 1:1 acid (HNO_3, for Mn, Fe, Ni and Cu and HCl for Zn and Cd) and diluting to 1 litre with DDW. To prepare the Cr(Ill) stock, 3.0460 g anhydrous $CrCl_3$ was dissolved in about 200 ml DDW and diluted to 1 litre with DDW. The lead stock was prepared by dissolving 1.598 g of lead nitrate, $Pb(NO_3)_2$, in 1% (v/v) HNO_3, and diluting to 1 litre with DDW, Nitrates were used because these ions have no/little affinity for metals. The pH was adjusted to 6.5 with 0.1mol/l sodium hydroxide. The interaction studies were carried out in batch tests.

1 g of soil was added to each of five sets of two bottles for one concentration of a metal, and 100 ml of the metal solution was added to each bottle in ten 250-ml PVC bottles. The bottle was shaken for 8 hours at room temperature in a reciprocating shaker at a rate of 100 forward and backward displacements per minute. The metal concentration was recorded every hour until the solution achieved equilibrium. The soil suspension was centrifuged at 2000 rpm for 10 minutes, then filtered through a 0.45-um membrane filter. The metals in the filtrate were determined using a Perkin-Elmer Analyst 100 AAS. Metal concentrations were measured twice for each feed concentration, and the findings were averaged. For each concentration, a control without soil was obtained to show that metal uptake due to the sorbent and not from other sources such as the container walls, centrifuge tube, etc. The uptake of metal ions on the soil at various initial feed quantities was calculated and presented in Table 2. Langmuir and Freundlich equations were used to analyse the data.

$\log a = \log k + 1/n \log c$ Freundlich equation

$c/a=1/Qb + c/Q$ Langmuir equation

where a (mg/g) is the metal sorbed per unit mass of soil (a= x/m where x mg of metal is sorbed on m grams of soil), k (mg/kg) and n are Freundlich constants related to adsorption capacity and adsorption intensity respectively, and Q (mg/g) and b (kg/g) are Langmuir constants related to adsorption capacity of soil and adsorption maximum (energy of adsorption) respectively. The Freundlich and Langmuir constants were calculated at four initial concentrations under optimal conditions. The effects of contact time and sorbent dose on sorption have also been investigated.

Table 2: Sorption percentage of Cr, Mn, Fe, Ni, Cu, Zn, Cd and Pb on 1 g of each of sandy loam and silty loamy soils in 100 ml metal solution at rpm 100 and pH 6.5 for 8 h

Metal	Soil type	Metal conc. In soil from deep horizon (mg/kg)	Metal conc. Applied (mg/l)	Adsorption (%)
Cr	Sandy loam	223.30	1 10 100 500	30.0 22.1 16.8 14.2
	Silty loam	175.00	1 10 100 500	40.1 34.0 28.3 23.6
Mn	Sandy loam	168.88	1 10 100 500	60.2 51.3 36.4 29.1
	Silty loam	167.20	1 10 100 500	80.4 69.2 58.1 42.5
Fe	Sandy loam	47476.00	1 10 100 500	44.9 34.2 29.5 24.1
	Silty loam	3560.00	1	60.2

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			10 100 500	51.4 43.0 36.5
Ni	Sandy loam	85.00	1 10 100 500	37.2 26.5 21.4 17.9
	Silty loam	125.50	1 10 100 500	50.5 41.2 33.8 27.6
Cu	Sandy loam	441.97	1 10 100 500	10.9 8.6 7.1 6.8
	Silty loam	423.89	1 10 100 500	15.2 10.8 8.4 7.4
Zn	Sandy loam	443.70	1 10 100 500	15.2 11.6 8.9 7.0
	Silty loam	476.00	1 10 100 500	20.3 15.0 12.4 10.9
Cd	Sandy loam	3.65	1 10 100 500	4.0 3.2 2.4 2.1
	Silty loam	5.20	1 10 100 500	5.0 4.5 3.9 2.4
Pb	Sandy loam	368.00	1 10 100 500	7.5 6.1 5.2 4.0
	Silty loam	288.00	1 10 100 500	10.4 8.2 6.5 5.1

Table 3: Analysis of the data using Freundlich and Langmuir equation for the sorption of Cr, Mn, Fe, Ni, Cu, Zn, Cd and Pb on sandy loam and silty loamy soil

Soil	Metal	Langmuir constant and R^2			Freundlich constant and R^2		
		Q, mg/g	b, l/g	R^2	K, mg/g	1/n	R^2
Sandy loam	Cr	4.9036	0.0320	0.9654	2.9236	0.0211	0.7954
	Mn	9.0524	0.0453	0.9542	4.0224	0.0301	0.6542
	Fe	8.7007	0.0404	0.9222	5.7047	0.0299	0.6122
	Ni	6.2491	0.0418	0.9788	4.2296	0.0276	0.8388
	Cu	2.7363	0.0207	0.9678	1.2353	0.0145	0.6678
	Zn	2.6622	0.0233	0.9321	1.1341	0.0162	0.7421
	Cd	0.9597	0.0133	0.9125	0.4547	0.0102	0.5615

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	Pb	1.9915	0.0124	0.9311	1.0035	0.0093	0.7311
Silty loam	Cr	9.6089	0.0212	0.9565	4.5059	0.0212	0.7454
	Mn	11.9651	0.0117	0.9842	7.5611	0.0059	0.6142
	Fe	12.9612	0.0416	0.9222	7.3642	0.0285	0.5812
	Ni	9.6929	0.0350	0.9892	5.3412	0.0278	0.8088
	Cu	2.6330	0.0304	0.9478	1.3210	0.0262	0.6478
	Zn	4.6454	0.0233	0.9621	2.3241	0.0146	0.7221
	Cd	1.4405	0.0477	0.9221	0.9108	0.0298	0.5815
	Pb	2.1564	0.0149	0.9482	1.0001	0.0089	0.7011

III. RESULTS AND DISCUSSION

The column method takes a significant amount of time and effort. So batch approach was employed because it was easier, faster, more reproducible, and less time consuming. The batch type system simulates saturated zone conditions in an aquifer. Their lengths and widths are often measured in meters, whereas their thickness is usually measured in meter [12]. Groundwater movement is caused by a hydraulic gradient that is determined by local geohydrological conditions and is usually measured in metres, but can also be measured in centimetres per day. Groundwater would have plenty of time to reach some sort of equilibrium with aquifer materials, whether it's pseudo or real. When compared to the amount of aquifer material, the volume of slowly moving groundwater is fairly significant. Based on this, it was determined, rather randomly, for the batch trials in the laboratory to use 1:100 solid : liquid ratios and a shaking rate of 100 forward and backward displacements per minute for 8 hours at room temperature. Only if the soil penetration rate is very slow batch experiments may be used to simulate soil-metal interactions. Metal retention by the soil of the unsaturated zone is likely to be lower.

It is observed in the present case that sorption of cations goes on increasing as their concentration decreases from 500 to 1 mg/L. The sorption percentage at all levels of Mn^{2+} is more that of all the cations under this study, and Fe^{3+} is about 10% more than that Ni^{2+} and of Pb^{2+} is more than that of the Cd^{2+} The sorption decreased in the order Mn >Fe> Ni>Cr> Zn> Cu> Pb> The leaching order should be reverse of it. Thus Cd^{2+} is the least strongly retained by the soils than to other toxic cations, and hence can pose a more serious problem of polluting groundwater with its extreme toxicity. Zn, Cu, Pb and Cd were found far more mobile than other four metals. These should, therefore, be properly monitored at all contaminated sites in Agra. An increase in metal concentration to weaker retention was observed. This indicates to possibility of groundwater pollution due to land filling.

Metals are better adsorbed on silty loam than on sandy loam due to increased organic matter clay, monmorillonite content, CEC, and surface area due to higher clay content. The degree of sorption in various soils is also determined by mineral elements such as $CaCO_3$, Fe_2O_3, Al_2O_3, and others. Adsorption dominates in sandy loam, while sorption dominates in silty loam, according to surface charge densities combined with sorption data. The primary components in silty loam samples appear to be sorption and ion exchange. It's possible that the great dominance of adsorption over sorption in sandy loam is attributable to the area's highly polluted groundwater[15]. Because the majority of the soil in Agra is sandy loam, all garbage containing heavy metals should be disposed of properly.

The sorption decreased in the order Mn> Fe> Ni> Cr> Zn> Cu> Pb> Cd. This is probably due to several factors such as ionic size, ionic potential, pH, metal reactivity, soil properties and environmental conditions. The sorption data do not fit to the Langmuir isotherm but very well fit to the Freundlich equation. This shows that the interaction of metals with soil is governed by more than one mechanism, i.e. sorption is due to adsorption, absorption and ion exchange all taking place simultaneously along with the reverse effect of desorption.

At pH 8.8, humic and fulvic acids form stable and soluble metal complexes. As a result of the fulvic and humic acid complexes, metals become more mobile and may seep out and damage water supplies. Clay soil keeps a high concentration of metals, whose mobility reduces as soil organic matter (OM) increases, owing to the development of clay-metal-OM complexes. As a result, metal pollution can be reduced by utilising a liner made of high clay soil and keeping the pH of the fill above 8.3.

Both agriculture and the environment benefit from the research. It introduces a new method for predicting, quantifying, and remediating (PQR) pollutants in soil, thereby safeguarding the aquifer. Increased OM, clay lining, waste treatment, pump and treat, surfactant enhanced subsurface remediation, wetland formation, zero-valent iron treatment, electrochemical remediation, and other remedial measures can be used with the authentic hydrochemical data obtained to work out suitable remedial measures to restore groundwater regime, such as increasing OM, clay lining, waste treatment, pump and treat, surfactant enhanced subsurface remediation, wetland formation,

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zero-valent iron treatment, electrochemical remediation, This study's findings could aid in the development of a hydrogeochemical model for more accurate prediction of pollutant fate and transport in landfills. The knowledge gained would be useful in developing future technology to liberate contaminants whose principal portions have degraded into non-recoverable forms, which, over geologic time, will eventually become non-recoverable forms.

IV. CONCLUSIONS

According to this study the adsorption, absorption, ion exchange and desorption take place simultaneously when heavy metals interact with loam soil. As a result, Cd^{+2} is less strongly retained by soils than the other hazardous cations, and so poses a greater risk of polluting groundwater due to its severe toxicity. The metals Zn, Cu, Pb, and Cd were shown to be significantly more mobile than the other four. As a result, all polluted sites in Agra should be closely monitored. Weeker retention was caused by an increase in metal content. This points to the probability of landfill-related groundwater pollution. Because the former contains higher organic matter, clay, monmorillonite concentration, CEC, and surface, metals are more absorbed on silty loam than on sandy loam. Because the majority of the soil in Agra is sandy loam, it is recommended that all wastes containing heavy metals be treated before being disposed of in loamy formation.

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