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## Study of the Mobility of Heavy Metals in Soil Amended with Sewage Sludge

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**Abstract:** Duplicate (250g) samples of air dried sewage sludge / mixtures in different percentages were placed in sintered glass to conduct a study of the mobility of heavy metals in soil amended with sewage sludge. The pH values of saturation extracts showed an increase in all sample mixtures up to 3<sup>rd</sup> month. The concentrations of nickel in the extracts were higher than those of cadmium and lead. There was a marked depletion in levels of metals with time, thus showing that the greatest release of metals occurred in the early stages after mixing probably due to the flushing out of the initial soluble forms already present in sludge. Another possibility could be that the decrease in metal concentrations with time could be due to the development of anaerobiosis.

**Key words:** Heavy metals, soil, sewage sludge, mobility, variation

### Introduction

Heavy metals released from sewage sludge are distributed throughout the soil system, while remaining in the soil solution as ions and inorganic or organic complexes are mobile for uptake by plants (Hooda *et al.*, 1997). This mobility and availability depends on several factors including soil texture and pH (Nouri, 1980 and Alloway, 1995). Mobile forms of metals released from sludge, which are not taken up by plant roots, may move down the profile and reach the water table. This pollution of groundwater may affect, surface waters and possibly potable water supplies. Changes can occur in chemical form and mobility of metals in the leachate, such as complexation of chelation, which are usually the results of variation in pH or reduction oxidation (Sims and Patrick, 1978; Essington and Mattigod, 1991). In situations where a high concentration of heavy metal ions exists in a leachate the pH is probably low and sorption/precipitation of these ions will be achieved by increasing the pH (Brallier *et al.*, 1996). Evidence for a high degree of retention of certain heavy metals in the top soil includes studies conducted in connection with the application of sewage sludge to agricultural soils (Keefer and Singh, 1986; Alloway and Jackson, 1991). Several workers have already investigated this topic and concluded that only relatively small amounts of metal were available for transport in the soil water immediately after sludge application (Slide and Kardos, 1977). Giordano and Mortvedt (1976) showed that, under excessive leaching conditions, movement of heavy metals in soil is somewhat greater from inorganic than from complexed sources found in sewage sludge.

The objectives of the experiments described here were to evaluate the relative quantities released and the movement of cadmium, nickel and lead on application of sewage sludge to various soils.

### Materials and Methods

The soil and sludge were collected and prepared in London in 1990. Duplicate 250 grams samples of air dried sewage sludge/ soil mixtures comprising 100, 90, 80, 50 and 0 per cent soil were placed in sintered glass membrane (No. 4) filter funnels and kept at the field capacity moisture by regular watering with deionized water. Soil solution was extracted by placing the funnel in a suction flask linked to a vacuum line. The first early extraction produced very little filtrate so an improved standardized procedure was adapted in which 40 ml

of deionized water was slowly added to the mixtures, 30 minutes before vacuum filtration. This filtrate was more realistically a "saturation extract" than a representative sample of soil solution. After filtration the pH values of the filtrates were determined and subsamples (>40 ml) were taken to dryness with concentrated nitric acid to destroy any soluble organic molecules prior to chemical analysis. For comparative purposes, small subsamples of the sludge/ soil mixtures, taken at the beginning and end of the experiment, were extracted with water and the metal concentrations determined in the extract (McLaren and Crawford, 1973). Twelve saturation extracts were obtained over a period of eight months (Tables 1 to 5). Measurements were also made of the redox potentials in the soil and sludge mixtures used. The technique involved using a platinum electrode and is subjected to considerable errors; however, it was useful as a qualitative indicator.

### Results

The pH values of saturation extracts given in Tables 1 to 5 showed an increase in all sample mixtures up to third month. In all cases the rise was relatively rapid in the first week and most pronounced for 100 per cent soil and 90 per cent soil/ 10 per cent sludge samples for all the metals studied. In the three sample mixtures in which soil predominated, there was a small decrease in pH after month 3, but the 100 per cent sludge sample showed a continuous increase from the beginning up to month 8. The 50 per cent sludge mixtures showed a decrease between months 3 and 6, followed by a slight increase between months 6 and 8. The data in Fig. 1 showed a rapid decrease in cadmium over the first month, which in most cases continued at a more gradual rate for the remaining months. One sample mixture (90 per cent soil/ 10 per cent sludge), showed a slight, but non-significant increase in cadmium, however the other mixtures showed a slight decrease. The concentrations of nickel in the extracts were higher than those of cadmium and lead. The nickel concentration showed a continuous decrease in all the samples except a slight increase after six months for (80 per cent soil/ 20 per cent sludge) and (90 per cent soil/ 10 per cent sludge). The release of lead into the filtrate was relatively rapid in first month, with the largest amounts being in 100 per cent sludge and 90 per cent soil/ 10 per cent sludge mixture. The rates of release of metals decreased continuously

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Table 1: Variation in metal concentrations and pH filtrate with time (100% soil) (metal values in  $\mu\text{g ml}^{-1}$ )

Total time	pH	Cadmium	Lead	Nickel
0	4.7	0.007	0.17	0.1
5 hour	5.2	0.008	0.13	0.09
1 day	6.3	0.009	0.12	0.09
4 days	6.5	0.01	0.09	0.08
11 days	6.6	0.008	0.04	0.07
18 days	6.7	0.004	0.04	0.06
25 days	6.9	0.002	0.03	0.04
1.5 months	7.0	0.0015	0.028	0.05
2 months	7.1	0.001	0.023	0.04
3 months	7.2	0.001	0.02	0.03
6 months	7.0	<0.001 *	0.015	0.024
8 months	7.0	<0.001	0.011	0.015

\*0.001 value is the limit of detection.

Table 2: Variation in metal concentrations and pH filtrate with time (90% soil + 10% sludge) (metal values in  $\mu\text{g ml}^{-1}$ )

Total time	pH	Cadmium	Lead	Nickel
0	4.6	0.12	0.25	1.35
5 hour	5.5	0.1	0.14	1.15
1 day	6.2	0.05	0.12	0.47
4 days	6.4	0.05	0.09	0.37
11 days	6.5	0.027	0.05	0.24
18 days	6.6	0.02	0.03	0.22
25 days	6.8	0.013	0.02	0.19
1.5 months	6.9	0.012	0.019	0.17
2 months	7.0	0.009	0.018	0.15
3 months	7.2	0.007	0.019	0.14
6 months	6.7	0.006	0.015	0.12
8 months	6.6	0.004	0.015	0.14

Table 3: Variation in metal concentrations and pH filtrate with time (80% soil + 20% sludge) (metal values in  $\mu\text{g ml}^{-1}$ )

Total time	pH	Cadmium	Lead	Nickel
0	5.6	0.13	0.20	3.0
5 hour	6.0	0.1	0.16	2.1
1 day	6.1	0.06	0.12	1.25
4 days	6.2	0.05	0.1	0.97
11 days	6.3	0.05	0.09	0.69
18 days	6.4	0.04	0.08	0.53
25 days	6.6	0.02	0.047	0.39
1.5 months	6.8	0.15	0.03	0.29
2 months	7.0	0.013	0.026	0.3
3 months	7.3	0.01	0.022	0.28
6 months	7.0	0.011	0.032	0.24
8 months	7.0	0.01	0.03	0.26

Table 6: water soluble metal concentrations in soil/ sludge mixtures ( $\mu\text{g ml}^{-1}$ )

Treatments	Cadmium		Lead		Nickel	
	Beginning	End	Beginning	End	Beginning	End
Soil (100%)	0.02	<0.01 *	0.1	<0.01	0.1	<0.01
Soil + Sludge (90%) (10%)	0.08	<0.01	0.25	<0.01	1.15	1.0
Soil + Sludge (80%) (20%)	0.1	<0.01	0.75	<0.01	4.2	2.3
Soil + Sludge (50%) (50%)	0.2	<0.01	1.0	<0.01	11.2	2.8
Sludge (100%)	0.3	<0.01	2.5	<0.01	34.3	5.4

\* 0.01 value is the limit of detection

Table 7: pH values of soil/ sludge mixtures\*

Treatments	pH at the beginning	pH after 4 months	pH after 8 months
Soil (100%) Soil + Sludge	6.3	6.8	6.6
(90%) (10%) Soil + Sludge	6.0	6.7	6.3
Soil + Sludge (80%) (20%) Soil + Sludge	5.8	6.6	6.3
Soil + Sludge (50%) (50%)	5.5	6.2	6.0
Sludge (100%)	5.3	6.0	6.1

\* Soil, sludge pH in  $\text{CaCl}_2$  solution.

Table 4: Variations in metal concentrations and pH filtrate with time (50% soil + 50% sludge) (metal values in  $\mu\text{g ml}^{-1}$ )

Total time	pH	Cadmium	Lead	Nickel
0	5.5	0.16	0.24	11.0
5 hour	5.6	0.12	0.2	6.6
1 day	5.7	0.1	0.16	4.1
4 days	5.8	0.08	0.12	2.43
11 days	5.9	0.05	0.11	2.42
18 days	5.9	0.025	0.1	1.98
25 days	6.0	0.02	0.06	1.79
1.5 months	6.0	0.019	0.055	1.6
2 months	6.0	0.018	0.04	1.29
3 months	6.2	0.017	0.038	1.23
6 months	6.0	0.015	0.04	0.8
8 months	6.1	0.01	0.035	0.63

Table 5: Variations in metal concentrations and pH filtrate with time (100% sludge) (metal values in  $\mu\text{g ml}^{-1}$ )

Total time	pH	Cadmium	Lead	Nickel
0	4.4	0.37	0.44	20.0
5 hour	4.9	0.36	0.40	14.0
1 day	5.0	0.23	0.39	10.0
4 days	5.2	0.18	0.35	9.3
11 days	5.3	0.16	0.30	8.9
18 days	5.4	0.08	0.20	7.0
25 days	5.5	0.05	0.12	5.3
1.5 months	5.6	0.03	0.11	3.8
2 months	5.7	0.025	0.08	3.0
3 months	5.8	0.02	0.05	2.1
6 months	5.9	0.018	0.04	1.4
8 months	6.0	0.008	0.04	1.0

for all samples up to 3 months, and then slightly increased except in 100 and 90 per cent soil where the rates of release continued to decrease. The samples of the mixtures taken at the beginning and end of the experiment, which were extracted with water, showed a marked depletion in levels of metals, with time (Table 6). Table 7 also shows the pH values of soil/ sludge mixtures at the beginning, middle and end of the experiment. The pH values at the beginning were the lowest, with the highest values in the middle part.

## Discussion

When sludge is applied to soil, it undergoes microbiological decomposition and releases heavy metals into the soil system over a period of time. Some decomposition and oxidation have already taken place before the sludge is applied to the soil and soluble forms of metals will already be present, these were probably flushed out at the beginning of the experiment. However, their amounts have been depleted with time and part of the fraction of metals released from the sludge are sorbed within the soil system. Thus it appears that the greatest release of metals occurred in the early stages after mixing and was probably due to the "flushing-out" of the initial soluble forms already present in sludge (Alloway, 1998). Very little bound metal would have been released from the sludge by microorganisms in this short time. In addition, sorption of mobile forms of metals within the soil system have occurred. Bradford *et al.* (1975) and Alloway and Jackson (1991) reported that the concentrations of metals were consistently greater in the extracts obtained from sludges than those obtained from soils. Microbiological activity would also help to cause, breakdown of organic metal-containing complexes in sludge, and possibly bring about the release of metals from soil state forms, such as sulfides, phosphates and carbonates. With the "flushing" of soil, bases are removed and the pH of saturation extract increases over the first three months before gradually decreasing. A similar trend was observed in the remaining soil, and sludge pH values. The soil solution became more diluted with time, therefore adsorption of divalent  $\text{Cd}^{2+}$ ,  $\text{Ni}^{2+}$  and  $\text{Pb}^{2+}$  ions was likely to be stronger. The solid phase of the soil releases heavy metals into solution partly by weathering of minerals and decomposition of organic matter, and partly by cation exchange (Essington and Mattigod, 1991). In addition to the increase in pH caused by flushing out of bases from soils, ammonium ions would probably start to increase as a result of microbiological activity on proteins in the sludge, and this would also increase the pH.

Redox potentials measurements in the soil and sludge mixtures used, showed that by four weeks after the commencement of the experiment, reducing conditions were found to exist in all treatments. The decreases in soluble cadmium, nickel and lead from sludge-soil mixtures with time were supported by the results of the water and ammonium acetate extractions which showed that the depletion was greatest in the mixtures with the highest proportions of sludge. It was also possible that the decrease in metal concentration with time could have been due in part to the development of anaerobiosis as was found by Bloomfield and Pruden (1975). However, it must be remembered that the laboratory conditions (warm and moist) may not necessarily be representative of fluctuating field conditions.

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