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LONG-TERM EFFECTS OF MUNICIPAL SEWAGE ON SOILS AND PASTURES

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ABSTRACT

Land application of municipal wastewater is widely practised worldwide as a means of treating wastes and obtaining a benefit from the water and nutrients by growing pastures, trees, and sometimes edible crops such as vegetables, fruit and fibre, etc. Melbourne in Australia has had such a 'sewage farm' at Werribee, about 40 km southwest of the city centre, since 1897. The Werribee Sewage Farm and is managed by Melbourne Water. The sewage, used to irrigate the mainly cattle grazed pastures, is made up of both domestic and industrial effluents.

Data on the chemical composition of the sewage sent to the Werribee Sewage Farm in the early decades could not be located, but data for the 1993-1995 period are assumed to be representative. Interest in changes in the soil due to irrigation developed in the fifties with a study of P accumulation (Aung Khin and Leeper, 1960) and in the seventies with research on heavy metals accumulation (Johnson et al., 1974; Evans et al., 1978; Melbourne Water data). There now exists a reasonable database for predicting trends in soil properties. Analysis of data collected over a period stretching from 1976 to 1993 shows that:

- heavy metals are generally immobilised in the top 30-40 cm of the soil and their concentrations are increasing slowly,
- heavy metals with known higher mobility, such as cadmium, have moved deeper into the soil than those with known low mobility,
- heavy metal concentrations in the forage are decreasing slowly, and
- soil pH is increasing slightly.

Other chemical changes in the soil may also affect the bio-availability of heavy metals to plants.

With continued application of sewage, assuming it is the same quality as the sewage applied till now, the concentrations of heavy metals in the topsoil will sooner or later exceed the criteria for contaminated soil (ANZECC-NHMRC, 1992; EPA(Vic.), 1995). However, from the point of view of forage quality, the long-term use of sewage for growing pastures appears to be sustainable.

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The questions that need to be addressed are what can we learn from this case study and what kinds of soil and pasture chemical analyses are needed to monitor these processes more satisfactorily?

SEWAGE MANAGEMENT AND LONG-TERM EFFECTS ON SOILS

Werribee is a dry location (annual rainfall 500-550 mm and pan evaporation 1400 mm) situated on Port Phillip Bay southwest of Melbourne. The topography is largely flat with minor undulations. The soils have formed from the weathering of the underlying basalt and from alluvium of mixed basaltic and sedimentary rock origin making up the Werribee River delta. The soils are mainly texture contrast soils (FAO, 1988: Calcic Luvisols; Vertic Luvisols; Soil Survey Staff, USDA, 1960: Rhodustalfs and Typustalfs, and vertic intergrades) and tend to have free calcium carbonate at depth of about 30 cm . The subsoils are medium to heavy clay, sodic and have a high shrink-swell capacity,. The pH increases from between 5 and 7 in the surface to 8-9 in the subsoil (Maher and Martin, 1952).

The locality is attractive for establishing a sewage irrigation scheme due to the low cost of the land and the high water demand in this rain shadow area. At the farm sewage is either treated in lagoons before discharge to the Bay (46%) or irrigated without prior biological treatment (54 %).

Infiltration in the alluvial soils is in the order of 20 mm per day. The clay soils that have formed in situ on the basaltic parent rock tend to have a lower permeability and infiltration capacity than the alluvial soils and as a result they are managed differently.

The total area of the sewage farm is about 10,900 hectares. Irrigation occurs on 5600 hectares of the land. Of this 4,200 hectares are devoted to shallow basins where the sewage is allowed to infiltrate into the more permeable alluvial soils (Land filtration). The remaining 1,400 hectares are used to treat the sewage by running it slowly through a grass cover on gently sloping land with the less permeable basaltic clay soils (Grass filtration). In addition about 1,375 hectares are used for oxidation lagoons and 2,800 hectares are reserved for dry grazing of livestock. The average weekly flow used for irrigation is 1734 ML. The average irrigation application is about 11.2 ML per hectare per year.

The chemical composition of the incoming sewage has been monitored every two weeks for a range of heavy metals, nutrients, salinity and pH. We have used the data for the 1993-1995 period. The heavy metal parameters are shown in Table 1 below.

Table 1 Heavy metal and total P concentration (mg/L) in raw sewage and annual loadings of the soil by irrigation for 1993-1995

	Cd	Cr	Cu	Fe	Hg	Ni	Pb	Zn	Total P
Mean Raw Sewage	0.11	0.587	0.253	2.28	0.002	0.09	0.218	0.77	10.8
Loading kg/ha.yr	1.77	9.45	4.07	36.71	0.32	1.45	3.51	12.40	174
Loading Moles/ha.yr	15.75	181.7	64.1	657.3	1.6	24.7	16.9	189.7	5615

Regular monitoring of the irrigated land and pastures by the responsible authority consisted of sampling the soil in both the land filtration and grass filtration areas at 0-10 cm and 10-

20 cm depth, and the herbage grown under both types of irrigation, twice a year. The wastewater flows were measured every two weeks and wastewater composition was measured monthly. This paper considers 17 years of data from 1976 to 1993.

As is to be expected, the heavy metals accumulated in the soil. This was noted by Johnson et al. (1974) and by Evans et al. (1978) for the same farm. Evans et al. (1978) took 15 core samples to a depth of 65 cm in irrigated and non-irrigated areas. Core samples were collected in 5 cm intervals to 30 cm, and then in 10 cm intervals which enabled a depth function of the heavy metal concentrations to be established. The highest increase in concentration of metals was always in the upper 5 cm, however copper and nickel also have an increase – a 'bulge' - at about 30-50 cm below the surface. They also calculated the mean rate of accumulation for the top 5 cm as shown in Table 2 below and Figure 1.

Table 2 Accumulation of heavy metals in the upper 5 cm (Evans et al., 1978)

Time interval	Mean rate of accumulation (top 5 cm) in kg/ha.year					
	Cd	Cr	Cu	Pb	Ni	Zn
1900-1976	0.06	4.3	1.4	3.2	1.0	7.8

Lawes (1993) also investigated the heavy metal accumulation and mobility within land filtration areas of the Sewage Farm. She found that Cd, Cu, Ni and Zn had accumulated significantly in the upper 25-30 cm of soil, while Cr and Pb accumulation appeared to have been restricted to the top 15 cm. Irrigation increased the extractability of Cd, Cr, Cu, Ni and Zn in Na_2-EDTA, particularly that of Cu, Ni and Zn. In some instances it appeared that even the pre-existing or 'native' metal content had become more mobile through irrigation at the farm, as in these cases 100% of total Cd, Cu, Ni and Zn present in irrigated soil was extractable with Na_2-EDTA.

The solubility of iron oxides in Na_2-EDTA was also increased as a result of irrigation, possibly due to the frequency and duration of anoxic conditions during flood irrigation, which may prevent the formation of crystalline Fe oxides, with the result that amorphous precipitates may dominate.

The pH of the soil below a depth of 7.5 cm had also increased by about 0.5 unit as a result of irrigation.

The longevity of the monitoring by Melbourne Water enables trends of heavy metal concentration in the soil to be constructed. In spite of the enormous variability from year to year, the best fitting linear correlations all show slow increases in concentration. The fact that all behave in the same predictable manner should be interpreted as indicating significant trends, despite the variability of the data. Figures 2 and 3 are illustrative of these trends.

Figure 1 Depth functions of total heavy metals and HCl-extractable heavy metals at Werribee Sewage Farm (Evans et al. 1978)

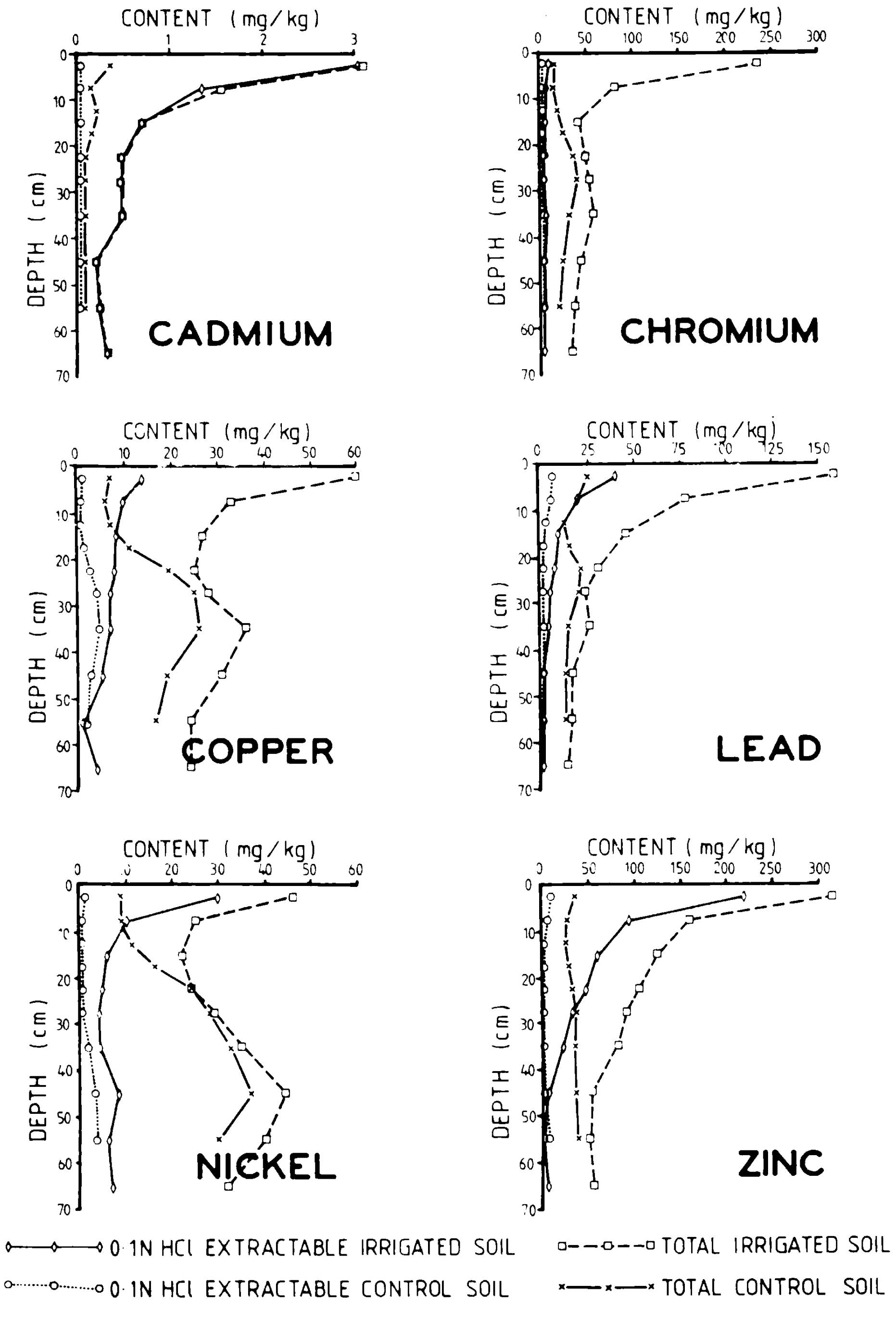
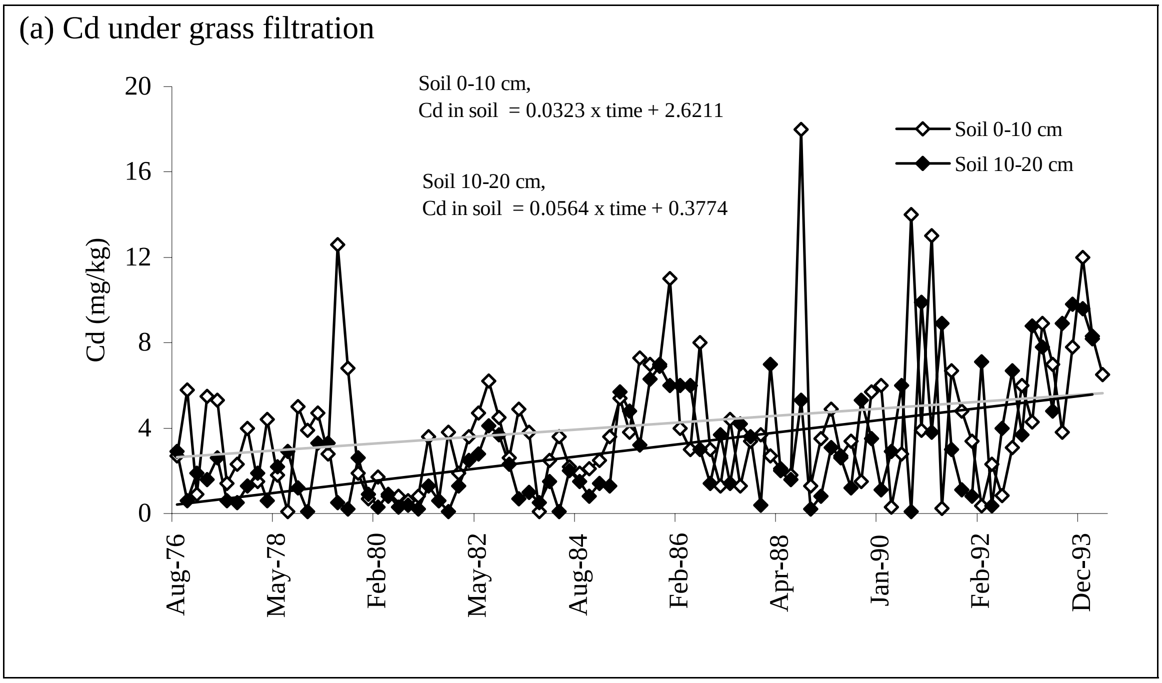


Figure 2 Accumulation of cadmium (Cd) in soil under grass filtration and land filtration at Werribee, from 1976 and 1993.



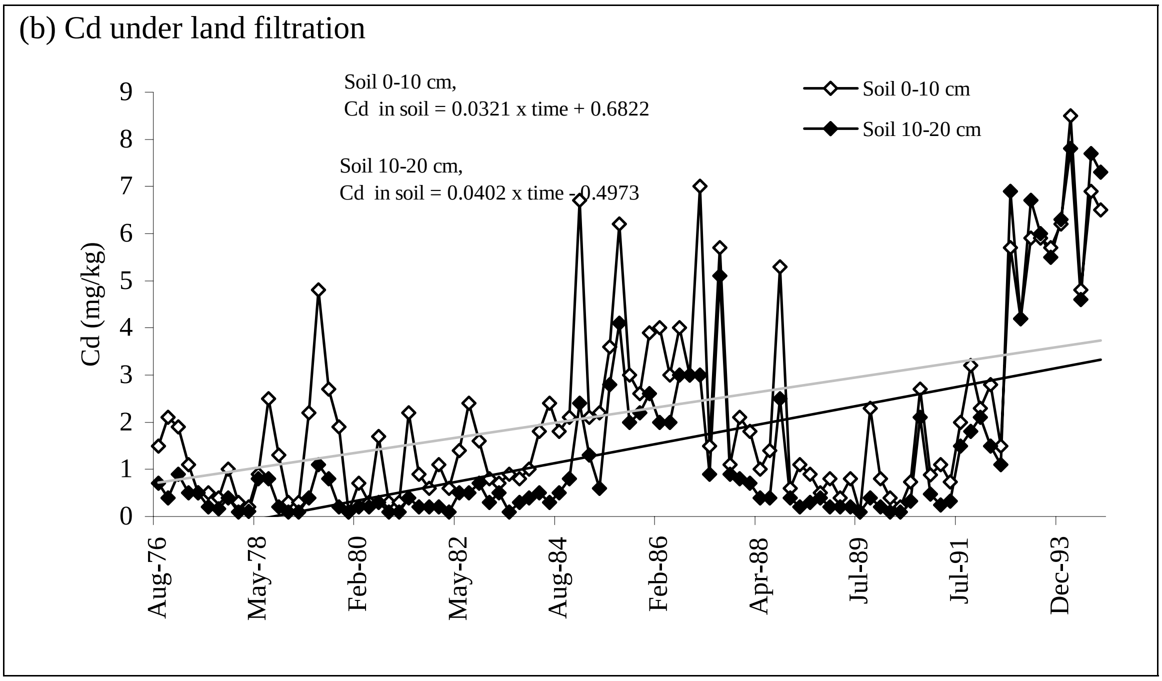
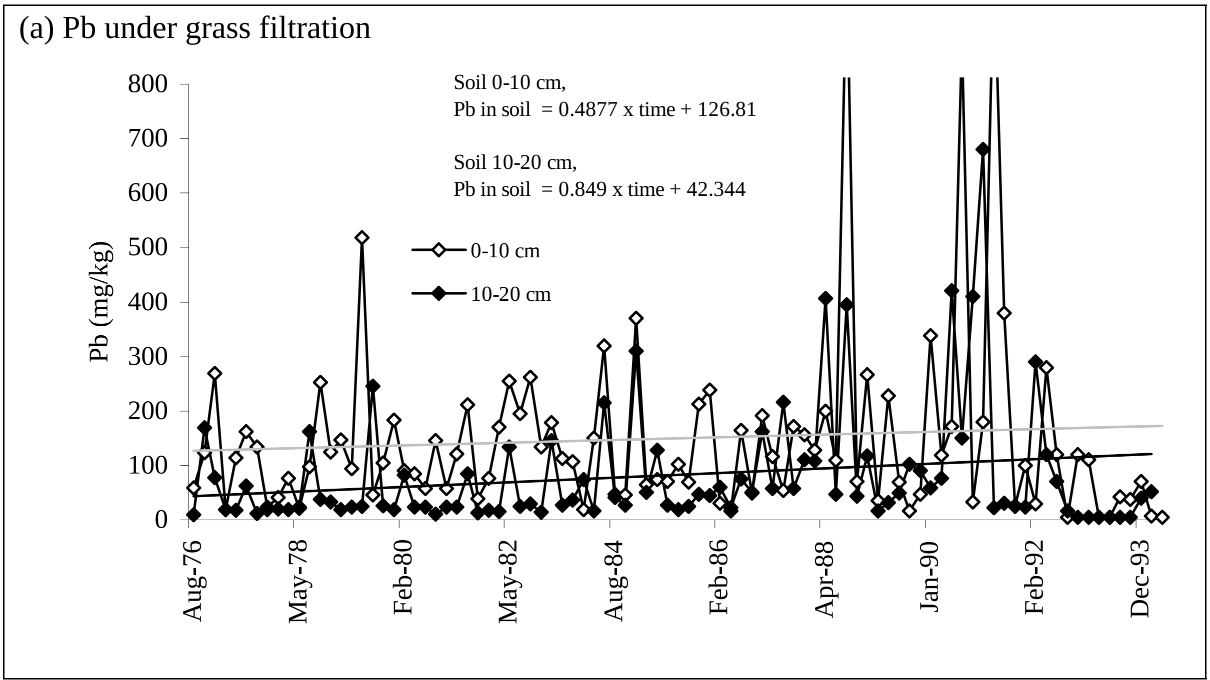
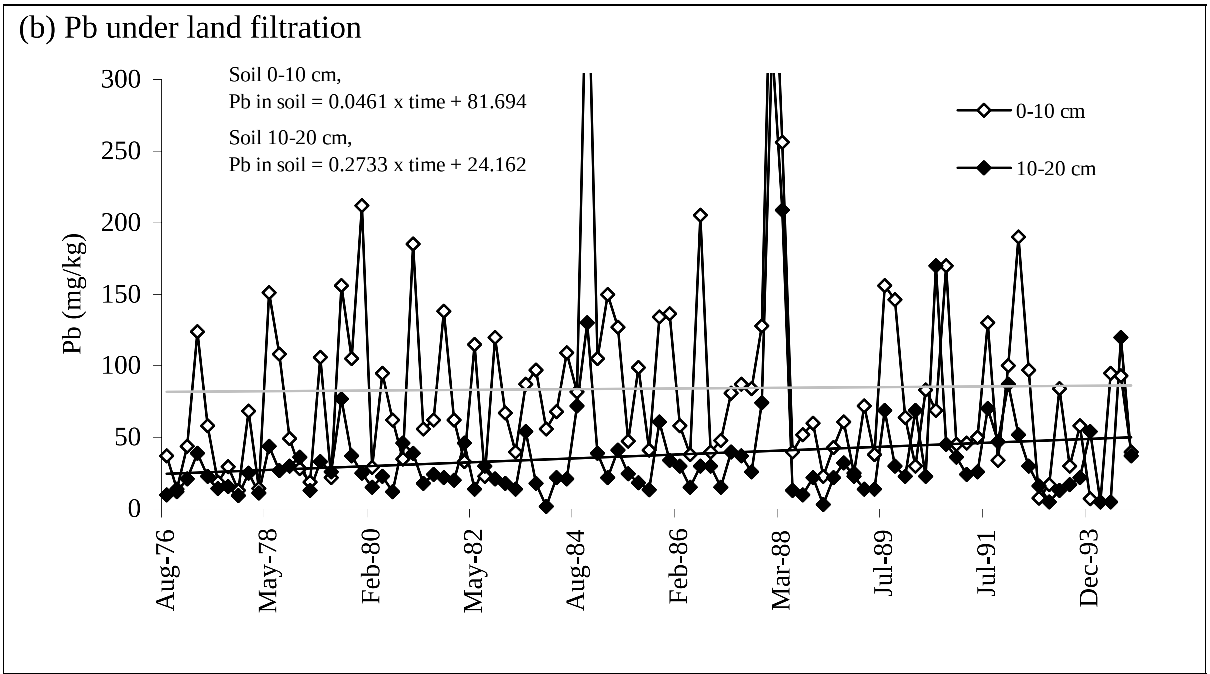


Figure 3 Accumulation of lead (Pb) in soil under grass filtration and land filtration at Werribee, from 1976 and 1993.





The high 'noise' of the data is probably due to the fact that heavy metal concentrations in soil can vary greatly over very short distances, so that, if only single soil samples are collected at any one time, the results will show the same variability. We assume that by taking a larger number of samples and compositing them the variability can be reduced and regressions will show greater statistical significance.

EFFECTS ON HEAVY METAL CONCENTRATIONS IN HERBAGE

Interestingly, the concentrations of heavy metals in the herbage harvested from the pastures show a general slow decrease over the period of observation. Due to improved wastewater management at source in Melbourne's industry the sewage should over time contain lower concentrations of heavy metals and this should also increase sustainability.

Evans et al. (1978) noticed that the difference in heavy metal concentrations between samples collected from the irrigated situation and non-irrigated situation, decreased down the food chain from soil to pasture to animal tissue (cow's liver), with the exception of copper. These trends are shown in Figure 4 and Figures 5 and 6, which exemplify the trends for the 17-year period of monitoring.

Figure 4 Heavy metal transfers from soil to pasture to animal liver (Evans et al., 1978)

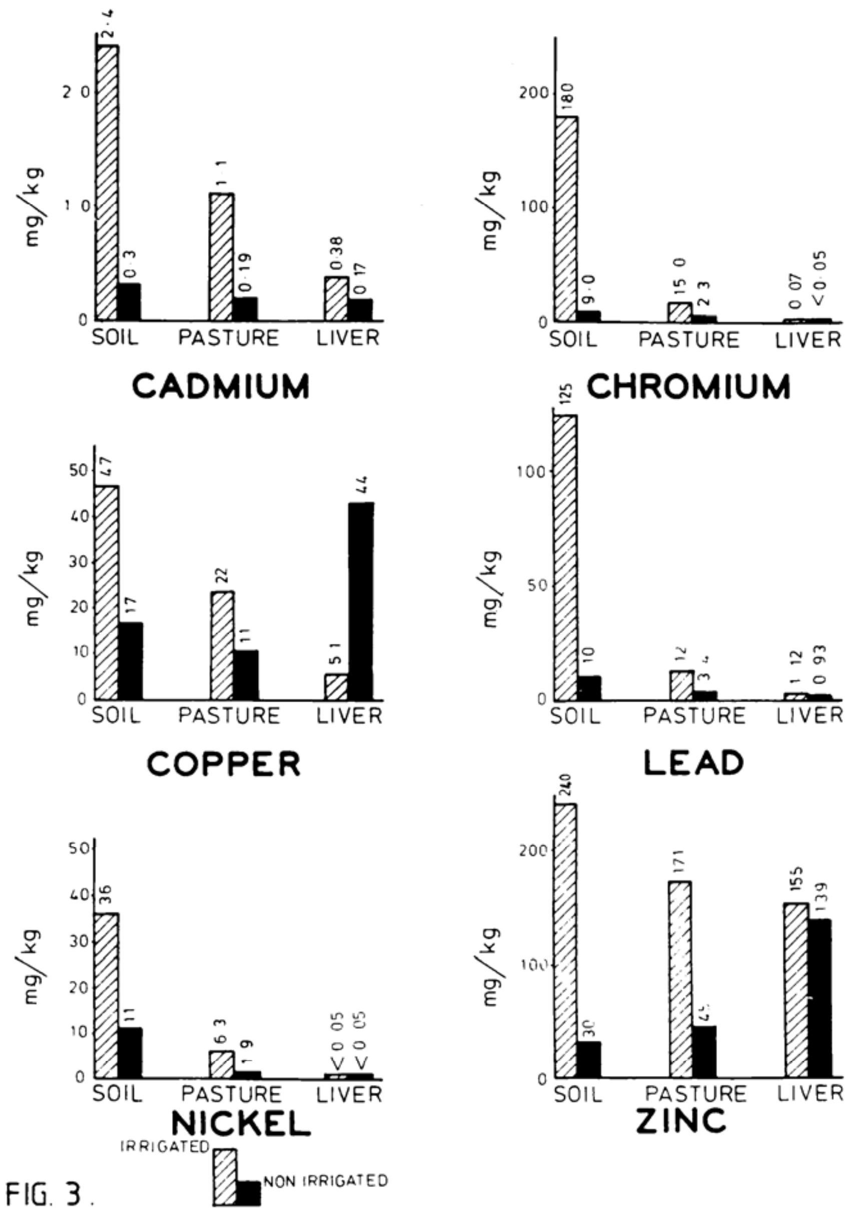


FIG. 3.
HEAVY METAL TRANSFER: SOIL→PASTURE→ANIMAL TISSUE

Figure 5 Cadmium (Cd) in herbage under grass and land filtration

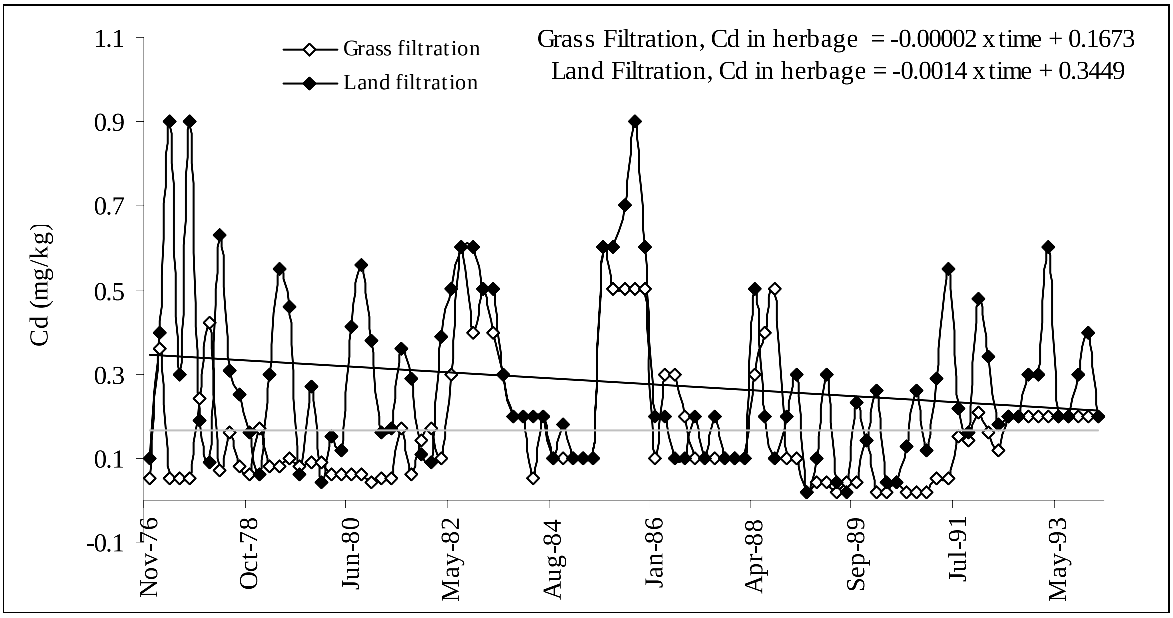
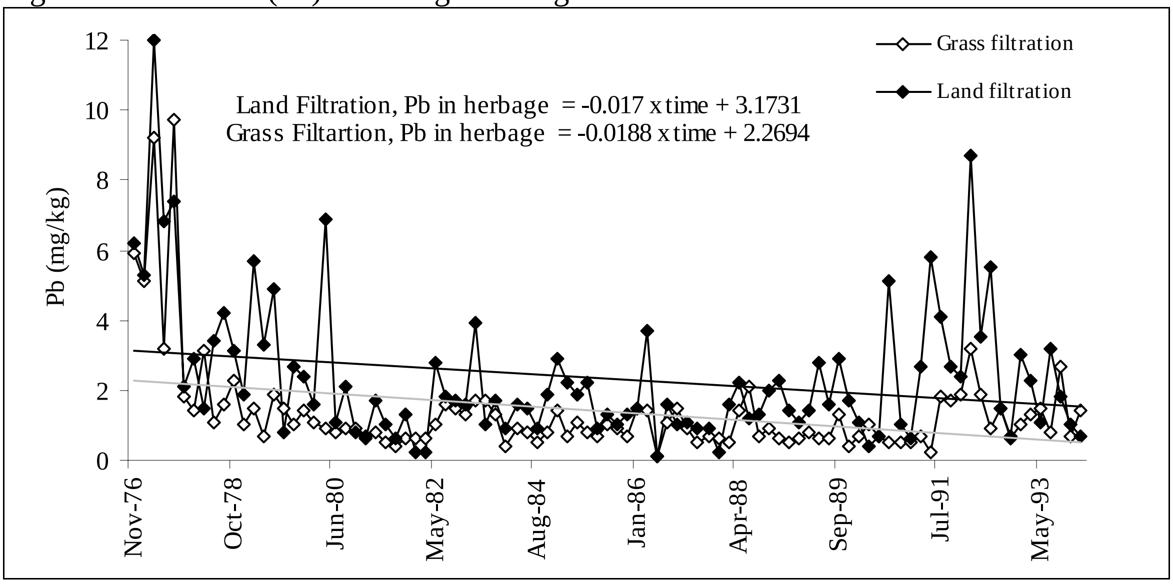


Figure 6 Lead (Pb) in herbage under grass and land filtration



RESULTS AND DISCUSSION

The decrease of metal concentration in the herbage suggests that in spite of the increasing concentrations in the irrigated soils there is decreased uptake.

Lawes (1993) has shown that the pH of the irrigated soil below a depth of 7.5 cm has increased by about 0.5 unit. For most of the metals in this study an increase in pH will induce a lowering of their concentration in the soil solution as follows from Table 3. How major this effect is would be difficult to determine in solutions with a plethora of other cations and anions.

Table 3 Ionisation constants (-log K) of the relevant heavy metals

	Cd	Cr	Cu	Fe	Hg	Ni	Pb	Zn
	$Cd(OH)_2$	$Cr(OH)_3$	$Cu(OH)_2$	$Fe(OH)_3$	$HgO+H_2O$	$Ni(OH)_2$	$Pb_2O(OH)_2$	$Zn(OH)_2$
K_T	14.4	30.2	18.8	38	25.4	14.7	14.9	15.5
K_1	9.5	11.8	12.1	16	15.2	11.3	-	10.5
K_2	4.9	8.4	6.7	9.4	10.2	3.4	-	5.0
K_3	-	10.0	-	11.7	-	-	-	-

Taking a simple cadmium hydroxide-water system as an example of the effect of increasing soil pH by 0.5 unit, the concentration of Cd^{2+} ions can be shown to be decreased by a factor of 10:

$K_T = 10^{-14.4} = [Cd^{2+}][OH^-]^2$ so that $[Cd^{2+}] = 10^{-14.4} / [OH]^2$

Therefore changing the pH from 6.0 to 6.5 increases the pOH from 8.0 to 7.5 and the nominator in the equation above changes from $[10^{-8}]^2 = 10^{-16}$ to $[10^{-7.5}]^2 = 10^{-15}$, a tenfold increase causing $[Cd^{2+}]$ to decrease by a factor of 10. We have assumed here that cadmium hydroxide can be treated as solid phase.

Unfortunately, calculations such as this may over-emphasize the responsiveness of solution concentrations to pH. Jeffery and Uren (1983) found that the concentration of zinc in the soil solution was highly responsive to pH but that of copper was not, and in the case of zinc it was less than would be predicted by the calculation used for cadmium above as the coefficients of pH in the regression equations below are much less than 1. Using an acid sandy loam amended with varying rates of lime and sphagnum peat, enriched with zinc or copper, they found the following linear relationships (Table 4).

Table 4 Empirically derived dependence of zinc concentration in the soil solution versus pH

Control	Limed	log (Zn) = 6.54 – 0.75 pH	$r^2 = 0.97$**
	Limed + Peat	log (Zn) = 4.82 – 0.50 pH	$r^2 = 0.96$**
Metal enriched	Limed	log (Zn) = 8.67 – 0.79 pH	$r^2 = 0.99$**
	Limed + Peat	log (Zn) = 6.93 – 0.60 pH	$r^2 = 0.99$**

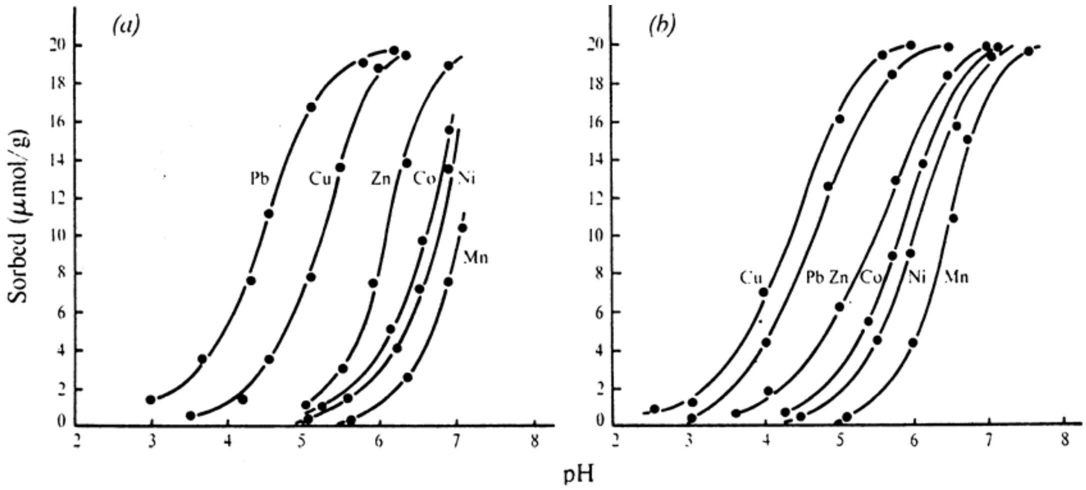
For copper the logarithm of the ratio (moderately labile and non-labile Cu/labile Cu) produced different but linear relationships with soil pH for the control and the amended soil.

Table 1 shows that there is a very large amount of iron added to the soil as a result of irrigation. Much of this iron could be in the form of suspended iron hydroxides or oxy-hydroxides. These may further react in the soil by forming new iron minerals and adsorb and occlude other heavy metals as shown in Figure 7 (McKenzie, 1980). The iron added to the soil by irrigation could well be in a reactive form and even immobilise pre-existing metals. Expressed as a loading of moles/ha.year the added iron far outstrips all the other added metals.

This process also takes place naturally in the geochemical environment of the Earth's surface, where soils rich in ferruginous materials can be shown to have accumulated a range of heavy metals and metalloids as well as a result of soil formation processes.

This hypothesis seemingly contradicts the finding by Lawes that iron oxides had become much more soluble as a result of irrigation. It is an unresolved question whether any iron oxides, having become crystalline during intensive soil drying periods would become mobile again in an anoxic 2-day irrigation period, and whether the increased extractability of iron with Na2-EDTA merely removed the freshly added iron.

Figure 7 Adsorption of heavy metals *v*. pH on iron oxides, 20 μmol/g added. (a) Hematite. (b) Goethite. After McKenzie (1980)



Finally, large quantities of P as phosphate are added to the soil by irrigation. Most phosphates of metals have very low solubility products (Table 5).

Table 5 Solubility products of various phosphates of heavy metals

	$Cd_3(PO_4)_2$	$CrPO_4$	$Cu_3(PO_4)_2$	$Fe(PO_4)$	Hg	Ni	$Pb_4O(PO_4)_2$
pK_{sp}	38.10	17.00-22.62	36.89	21.89	?	?	65.17
	$Pb_3(PO_4)_2$	$Pb_5(PO_4)_3Cl$	$Pb_5(PO_4)_3OH$	$Zn_3(PO_4)_2$			
pK_{sp}	44.60	84.40	76.80	32.04			

In conclusion there are three reasons why the metals appear to show decreasing bio-availability over time under sewage irrigation. These are: increasing pH lowering metal solubility; precipitation occurring with newly forming iron minerals; and precipitation occurring as insoluble phosphates. In the long run, decreasing loadings may also help.

Other metals like copper, nickel and manganese may have moved deeper into the soil profile as more mobile organic complexes or because the high BOD reduced their oxides, at least periodically, and made them more mobile.

It is difficult to know if these hypothetical effects continue over time ad infinitum or what an appropriate time scale could be. However, evidently over the century that the Werribee Sewage farm has been operated, it has proved to be sustainable and safe.

RECOMMENDATIONS

Monitoring of lands irrigated with sewage or other wastewaters should include measuring both the contaminants and, importantly, those parameters that are known to affect the solubility and availability of heavy metals. These parameters include pH, salinity (especially chloride), phosphate, iron and manganese, both in the added effluent and in the soil itself. In particular, the behaviour of iron, manganese and phosphorus in the soil and the effects of intermittent anoxic conditions should be researched in depth.

Analysis of the sustainability of the system should also include consideration of various land use scenarios that may occur after the wastewater irrigation has ceased, and their potential effect on soil chemistry.

All sampling being conducted as part of such monitoring should be based on taking many samples in an area and compositing these to reduce variability related to high spatial variation in the soils.

ACKNOWLEDGEMENTS

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