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# Heavy-metal uptake by crops from polluted river sediments covered by non-polluted topsoil

## *II. Cd-uptake by maize in relation to root development*

M. van Noordwijk, W. van Driel, G. Brouwer and W. Schuurmans
*DLO Research Institute for Agrobiology and Soil Fertility (AB-DLO), P.O. Box 129, 9750 AC Haren, the Netherlands*

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### Abstract

Cadmium uptake by maize from polluted river sediments covered with a clean top layer of variable thickness is discussed in relation to root distribution. Two pathways for uptake are distinguished: roots penetrating the contaminated layer or contaminants moving into the root zone. Relative Cd uptake proved to be roughly proportional to the fraction of total root length found in the contaminated layer. A deeper water table induced a deeper root development and more Cd uptake for a given thickness of clean topsoil. A model based on exponential decrease of root length density with depth is acceptable as first approximation only. Little or no evidence was found for contaminants moving into the root zone during the ten years of the experiment.

### Introduction

When a soil is too contaminated to grow crops which comply with public health standards, the simplest and cheapest solution often seems to cover such soil with less contaminated topsoil. This way of "covering up" problems obviously does not provide a real solution, but if the pollutants do not move into the root zone and the roots do not enter into the polluted soil, it may lead to a situation where all legal requirements are met without direct health risks.

The topsoil depth required to meet standards of permissible crop contamination is important in the economic evaluation of this option. Model calculations to evaluate a wide range of situations should be based on knowledge of underlying processes. The simplest assumption about root distribution is to expect an exponential decrease of root length density with depth (Gerwitz and Page, 1974). Considerable deviations from such a pattern can occur, however, and actual root length density in the subsoil can be both higher and lower than expected from such a description.

Two pathways exist for contamination of the crop: direct uptake from the contaminated zone and indirect uptake, after transport of contaminants into the root zone. The relative importance of both pathways depends on the nature of the contamination, on soil conditions in the topsoil, in the contaminated subsoil and their interface, on the crop, and on the water balance of the soil. Here we will evaluate the contribution of both pathways to Cd uptake by maize from a polluted river sediment (harbour sludge), covered by various depths of an uncontaminated clay soil. Compared to a range of other crops, maize requires a rather thick layer of clean topsoil in this situation (Van Driel et al., 1995). In the tenth and last year of a long-term experiment soil samples were taken to evaluate upward transport of metals and root distribution was measured to check the possible significance of the direct pathway.

For the root observations the following specific questions were formulated:

1. Is maize root distribution influenced by the depth at which the transition of topsoil to harbour sludge

*Table 1.* Treatments selected for detailed analysis and number of replicate samples taken; the topsoil and harbour sludge had similar texture and composition, except for soil pollutants; in series 2 an intermediate layer of coarse sand was used; further details are given by Van Driel et al. (1995)

| Experiment | Code | Topsoil | Intermed. | Harbour sludge | Water table | Core break | Calibration | Root maps |
|---|---|---|---|---|---|---|---|---|
| | | (m) | | | | No. of samples | | |
| Series 1 | A | 0 | 0 | 0.85 | > 1 | 100 | 10 | 2 |
| | C | 0.4 | 0 | 0.45 | > 1 | 100 | 10 | 2 |
| | E | 0.7 | 0 | 0.15 | > 1 | 100 | 10 | 2 |
| | F | 0.85 | 0 | 0 | > 1 | 100 | 10 | 2 |
| Series 2 | C | 0.25 | 0.15 | 0.45 | > 1 | 100 | 10 | 2 |
| | E | 0.25 | 0.45 | 0.25 | > 1 | 100 | 10 | 2 |
| Series 3 | A | 0 | 0 | 2.0 | > 2 | 90 | 10 | - |
| | B | 0.4 | 0 | 1.6 | > 2 | 30 | 10 | - |
| | C | 0.8 | 0 | 1.2 | > 2 | 45 | 10 | - |
| | D | 1.2 | 0 | 0.8 | > 2 | 30 | - | - |
| | F | 2.0 | 0 | 0 | > 2 | 105 | 10 | - |
| Total | 11 | | | | | 900 | 100 | 12 |

occurs or by the presence of an intermediate sandy layer?

2. Do roots accumulate at the edge of the 1 $m^2$ plots and reach the subsoil through this artificial pathway?
3. Is root distribution influenced by the difference in depth of water table between an experiment with topsoil layers up to 1 m and a raised experiment with up to 2 m of clean topsoil above the groundwater table?
4. Does an exponential decrease of root length density with depth provide an adequate description?
5. Is Cd uptake related to the root length in the harbour sludge?

Several mechanisms for transport of metals in the soil profile have to be considered (Van Driel and Nijssen, 1988), the relative importance of which may differ between situations:

**a.** mass movement of soil, e.g. by faunal activity,

**b.** mass flow of dissolved metals with water; a net leaching is to be expected, but periods of capillary rise can induce upward transport as well,

**c.** diffusion of metals in soil solution,

**d.** uptake by and redistribution in plants, leading to metal in root residues throughout the profile.

By measuring metal distribution in the profile as total content and in the soil solution, the relative importance of these mechanisms can be assessed: for mechanism **a** changes in total metal content are to be expected, for **b**, **c** and **d** primarily changes in dissolved metal content, possibly followed by precipitation; for mechanism **b** and **c** a gradual decrease in metal content upwards from the polluted layer, and for **d** a metal distribution proportional to root biomass, with the highest concentration in the top layers.

## Material and methods

Details of the experimental setup are described in part I (Van Driel et al., 1995). Eleven treatments were selected for more detailed sampling (Table 1). Topsoil consisted of a clay soil similar in texture and chemical fertility to the harbour sludge. In some profiles an intermediate layer of coarse sand separated harbour sludge and topsoil.

### *Core-break method*

To investigate root distribution with depth in eleven artificial soil profiles the "quick and dirty" core-break method was used (Anderson and Ingram, 1993). Cylindrical cores were broken in the middle and the number of roots protruding from the soil were counted. Results for both halves were added and expressed as root intensity, $N_h$, i.e. the number of intersections per $m^2$ of a horizontal plane. Equal numbers of samples were taken in and between crop rows, in two replicate plots per treatment. Two persons counted one half of the samples for each plot. A correction factor of 1.95 was found to be necessary to obtain comparable results for the second person.

To estimate root length densities $L_{rv}$ (km $m^{-3}$) from $N_h$, a calibration factor was derived from five samples per plot. In these samples, the soil was infiltrated with sodium-pyrophosphate overnight under vacuum and roots were washed free over a fine sieve (mesh size 0.3 mm). Root length was measured by the line intercept method (Tennant, 1975). The calibration factor combines experimental errors and effects of preferential orientation of roots.

*Root mapping*

Root distribution in a vertical plane was studied by the root mapping technique. After removing the concrete wall on one side of the small plots, a profile was smoothened at 50 mm from the edge of the plot. The soil was covered with a polythene sheet and all roots were mapped, as well as the layering of the profile. Subsequently the number of root intersections was counted for a 50 × 50 mm grid and expressed as $N_v$, root intensity in a vertical plane. Corrections were made where actual layering of the profile differed (maximum 30 mm) from the nominal one.

In the profile observations, specific attention was given to root morphology at the transition of topsoil to harbour sludge and to possible edge-effects in the small plots used. As a considerable difference in root development was noted between two replicates of the treatment with a 0.45 m sand layer, bulk density of the sand was measured with standard ring samples. No difference was noted (average bulk density 1.392 Mg $m^{-3}$).

*Chemical soil analysis*

The eleven mini-plots (Table 1) were sampled by taking core samples from the wet soil at 4–7 depth intervals (0–20, 20–40, 40–60, 60–80 cm (series 1 and 2) and 0–20, 20–40, 40–60, 60–80, 80–100, 100–140 and 140–180 cm (series 3). Per treatment for each depth interval 20 sub-samples were mixed to obtain one sample representative for the depth interval of the relevant mini-plot.

A portion of each sample was air-dried at 40°C for the determination of hot-acid-extractable metal concentrations. Soil solutions were extracted from approximately 100 $cm^3$ field moist soil samples by centrifugation at 20,000 N $kg^{-1}$ using an extraction cup similar to that described by Edmunds and Bath (1976) with the procedure developed by Del Castilho et al. (1993). Heavy-metal soil analyses were carried out in duplicate following the methods of Vierveijzer et al. (1979). Samples were wet-ashed with $HNO_3$ (Cd) or $H_2SO_4$-$HNO_3$ (Cu, Zn). Cu and Zn were measured in the water phase; Cd, after liquid-liquid extraction, in methyl-isobutylketone (MIBK), with flame atomic absorption spectrometry. In the acidified soil-solution samples Zn was analyzed using flame atomic absorption spectrometry, using standard additions and acid ammonium sulphate or nitric acid solutions as matrix modifiers. Cd and Cu were measured using electrothermal atomic absorption spectrometry (Del Castilho et al., 1993). Further details on the standard deviations of the analyses are given by Van Driel et al. (1995).

## Results

*Edge effect in root distribution*

Figure 1 shows contours of root intensity in a vertical plane, averaging data for two sides of 12 root maps. Highest root intensities were observed in between two plants at about 0.2 m depth. In the topsoil a pattern can be noted radiating from the plant. No specific accumulation of roots close to the concrete edge of the plot was noted. Statistical tests confirmed that root intensity close to the edge did not differ significantly from that in the middle of the map. No indications were obtained that roots penetrated deeper soil layers after creeping along the concrete wall, so that in this respect the experiment may be considered valid for unconfined field conditions as well.

*Calibration factors for root length density*

Results for root maps and for the core-break method can both be expressed as number of intersections per unit area (root intensity, $m^{-2}$). In both cases a similar trend with depth was noted, but data for the core-break method were consistently higher. Different results can

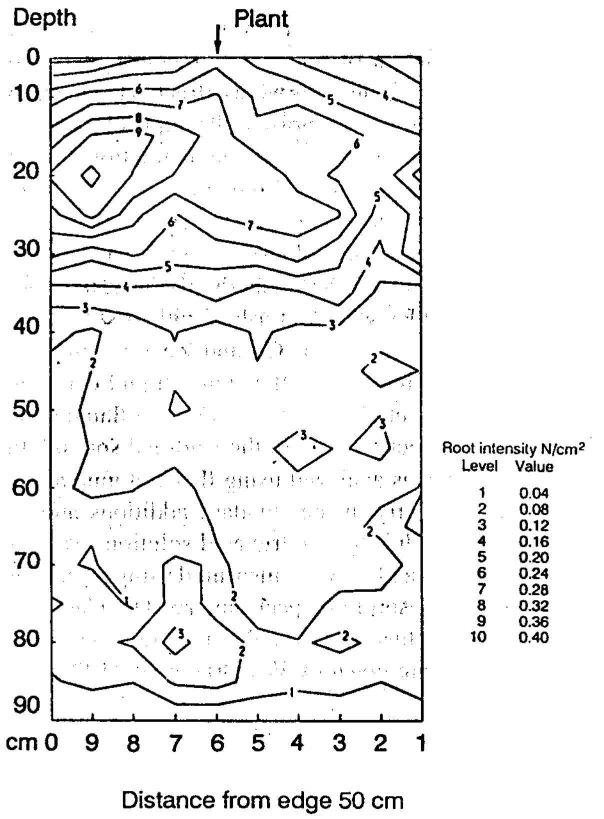


*Fig. 1.* Contour plot of root intensity (number of intersections per unit map area, decreasing from 0.4 to 0.04 intersections per cm²), in a vertical profile wall averaged over two sides of 12 maps.

be due to a preferential orientation of roots and to experimental errors. When data for all profiles and depths were combined, the following relation was found between $N_h$, root intensity in a horizontal plane (core-break method), and $N_v$, root intensity in a vertical plane (root map):

$$N_h = 416 + 3.351 N_v,\ r^2 = 0.656 \qquad (1)$$

The intercept in this regression equation was not significantly different from 0 but the slope was ($p < 0.01$). The ratio $N_h/N_v$ was not significantly influenced by depth or soil profile. If preferential orientation would cause the $N_h/N_v$ to differ from unity, a higher ratio would be expected for deeper layers in the profile.

Table 2 summarizes the regression analysis of root length density on root intensity observed with the core-break method. The expected relation is of the type $L_{rv} = 0.002\ X\ N_h$, where the calibration factor X differs from

*Table 2.* Regression lines for root length density $L_{rv}$ (km m$^{-3}$) and $N_h$ (m$^{-2}$), and for root weight density $D_{rv}$ (g m$^{-3}$) and $N_h$, for two regression models. For all parameter estimates statistically significant differences from 0 are indicated: ** ($p < 0.01$), * ($0.01 < p < 0.05$) and NS ($p > 0.05$)

| | $L_{rv} = a + b\ N_h$ | | | $L_{rv} = 0.002\ X\ N_h$ | |
|---|---|---|---|---|---|
| Depth (m) | a | b | $r^2$ | X | $r^2$ |
| 0–0.9 | 3.38** | 0.00133** | 0.476 | 0.930** | 0.354 |
| 0–0.1 | -2.60$^{NS}$ | 0.00186$^{NS}$ | 0.347 | 0.770** | 0.413 |
| 0.1–0.9 | 3.55** | 0.00136** | 0.522 | 0.975** | 0.359 |
| 0.1–0.5 | 3.65** | 0.00133** | 0.504 | 0.929** | 0.389 |
| 0.6–0.9 | 3.35** | 0.00147** | 0.471 | 1.105** | 0.265 |
| | $D_{rv} = a + b\ N_h$ | | | $D_{rv} = c\ N_h$ | |
| | a | b | $r^2$ | c | $r^2$ |
| 0–0.9 | 10** | 0.0079** | 0.524 | 0.0095** | 0.492 |
| 0–0.1 | -18$^{NS}$ | 0.0091$^{NS}$ | 0.419 | 0.0068** | 0.455 |
| 0.1–0.9 | 10** | 0.0086** | 0.580 | 0.0102** | 0.545 |
| 0.1–0.5 | 13$^{NS}$ | 0.0075** | 0.488 | 0.0094** | 0.451 |
| 0.6–0.9 | 5$^{NS}$ | 0.0116** | 0.694 | 0.0127** | 0.688 |

1.0 for preferentially oriented roots (Van Noordwijk, 1987) and under the influence of observation errors.

Regression analysis showed that a model of the form $L_{rv} = a + b\ N_h$ gives a slightly better fit and usually a statistically significant intercept a. For the present purpose we preferred the model $L_{rv} = 0.002\ X\ N_h$, however, as the first model overestimates root length density in zones where $N_h$ is (close to) zero. For the top 0.1 m a different regression equation was found than for the deeper layers. For the zones below 0.1 m the calibration factor X was approximately 1.0, indicating no preferential root orientation. The $N_h/N_v$ ratio was thus probably mainly caused by differences in observation method. With the core-break method roots tend to break not exactly in the plane of observation, so they stick out on one of the two sides and are better visible than on the profile wall which is smoothed before making a root map.

Root weight density, $D_{rv}$, was slightly better correlated with root intensity than with root length density, indicating that thin roots are more easily overlooked than thick ones. The ratio of root weight and $N_h$ increased with depth. Averaged for the whole profile, the specific root length (length per unit dry weight) was about 200 m g$^{-1}$, which is a normal value for maize (Van Noordwijk and Brouwer, 1991).

*Table 3.* Logarithmic regression equations of root intensity ($N_h$, $m^{-2}$) and root length density ($L_{rv}$, km $m^{-3}$) on depth H (m). $H_s$ = depth below which soil consists of harbour sludge. $L_{ra}$ = root length per unit soil surface area, is estimated as -a/b. $L_s^*$ = 100 ($e^{-b\ H_S}$). $L_s$ = observed percentage of root length in harbour sludge. $L_T$ = percentage of root length in topsoil = 100 - $L_s$, except for series 2 with an intermediate sand layer; in series 2 C and E it was 43.2 and 11.3%, respectively

| Code | | $H_s$ (m) | $\log(N_h) = a - b\,H$ a | b | $r^2$ | $\log(L_{rv}) = a - b\,H$ a | b | $r^2$ | $L_{ra}$ (km $m^{-2}$) | $L_s^*$ (%) | $L_s$ (%) |
|---|---|---|---|---|---|---|---|---|---|---|---|
| 0 < H < 1.5m | | | | | | | | | | | |
| Series 3 | A | 0 | 10.068 | 4.591 | 0.599 | 3.762 | 4.513 | 0.589 | 9.54 | 100 | 100 |
| | B | 0.4 | 9.604 | 4.385 | 0.535 | 3.298 | 4.307 | 0.526 | 6.28 | 18 | 37.8 |
| | C | 0.8 | 9.551 | 4.025 | 0.601 | 3.248 | 3.954 | 0.592 | 6.50 | 4.2 | 16.1 |
| | D | 1.2 | 9.463 | 4.683 | 0.619 | 3.159 | 4.608 | 0.612 | 5.11 | 0.4 | 0.6 |
| | F | - | 8.917 | 3.847 | 0.446 | 2.623 | 3.781 | 0.439 | 3.64 | 0 | 0 |
| 0 < H < 0.9m | | | | | | | | | | | |
| Series 1 | A | 0 | 8.909 | 4.494 | 0.252 | 2.592 | 4.356 | 0.241 | 3.07 | 100 | 100 |
| | C | 0.4 | 9.173 | 3.870 | 0.236 | 2.837 | 3.705 | 0.220 | 4.61 | 18 | 26.3 |
| | E | 0.7 | 9.326 | 2.631 | 0.175 | 2.994 | 2.473 | 0.155 | 4.34 | 18 | 11.5 |
| | F | - | 9.433 | 4.885 | 0.345 | 3.092 | 4.706 | 0.325 | 4.68 | 0 | 0 |
| Series 2 | C | 0.4 | 9.151 | 2.463 | 0.189 | 2.815 | 2.301 | 0.167 | 7.26 | 40 | 39.4 |
| | E | 0.7 | 9.941 | 8.883 | 0.554 | 3.601 | 8.696 | 0.544 | 4.21 | 0.2 | 4.5 |

*Root distribution with depth*

Root intensities were now transformed into estimated root length densities. Table 3 gives parameters for an exponential decrease of root length density with depth, H. With the deeper groundwater table (series 3), such a model gave an acceptable description, accounting for about 50% of all variation in the core-break results. Combining data for five profiles the best fit is:

$$L_{rv} = 24.1e^{-4.17H}, r^2 = 0.529 \qquad (2)$$

For the shallower groundwater tables (series 1 and 2), an exponential description is much less convincing, as it accounts for only 15–30% of the variation, except for series 2 profile E, with the 0.45 m thick sandy intermediate layer. For series 1 and 2, except 2E, the best fit is:

$$L_{rv} = 17.7e^{-3.51H}, r^2 = 0.214 \qquad (3)$$

From the exponential model an estimate of the total root length per unit soil surface area, $L_{ra}$, can be obtained by integration:

$$L_{ra} = \int_0^\infty L_{rv}dH = \int_0^\infty ae^{-bH}dH = -\frac{a}{b} \qquad (4)$$

The highest $L_{ra}$ value was found for the deep profiles consisting of only harbour sludge.

The fraction of root length, $L_s^*$, below a depth $H_s$ can be obtained as:

$$L_s{}^* = e^{b\,H_s} \qquad (5)$$

In Table 3 the estimated $L_s^*$ obtained from the exponential model are compared with the observed fractions $L_s$. Considerable differences indicate that the exponential model is not a safe basis for predictions of the root length present in the harbour sludge.

Figures 2 and 3 give the observation points as well as exponential curve fits for root length density for each profile. Again, no specific response to the transitions in the profile is evident, except for the sandy layers in series 2. In the sand a low root length density was found, but the few roots which reached the harbour sludge branched abundantly in this zone, resulting in a root length density in the sludge similar to that without intermediate sand layer. The deeper groundwater table in series 3 led to a deeper root development. A topsoil layer of 0.4 m contained 74% of the roots in series 1 and only 62% at the deeper groundwater table in series 3.

*Cadmium uptake*

Cd concentrations in aboveground biomass can be expressed as relative amounts by subtracting the Cd

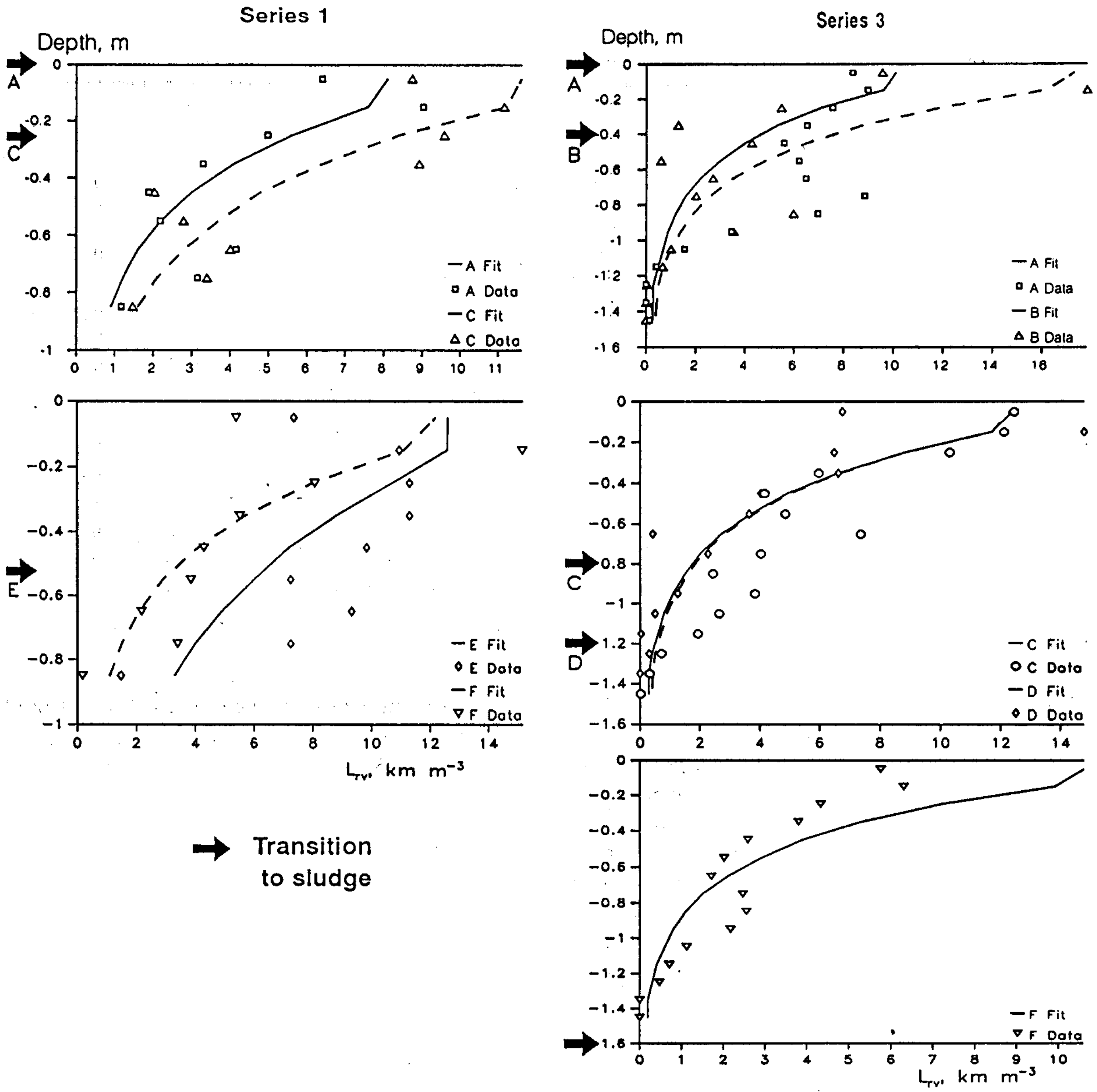


*Fig. 2.* Root length density $L_{rv}$ as a function of depth H for the profiles (of series 1 and 3 see Table 1) without an intermediate sandy layer. Observation points are given as well as an exponential model fitted on $N_h$. Arrows indicate the transition from topsoil to harbour sludge for each treatment.

concentration in the unpolluted control and dividing by the Cd concentration in the crop grown without unpolluted topsoil (again minus the control value). Figure 4 relates this relative Cd content (Van Driel et al., 1995) of maize to the relative amount of roots in the polluted zone, $L_H$. Most of the data points are slightly (about 5%) above a 1:1 line, indicating that Cd uptake is a bit higher than to be expected from root development in the subsoil. This effect might have been caused by a slight underestimation of the Cd content of crops grown in topsoil only. As we do not have replication at this level, a statistical test can not be made. The positive intercept with the Y-axis of about 5% is probably within the error range.

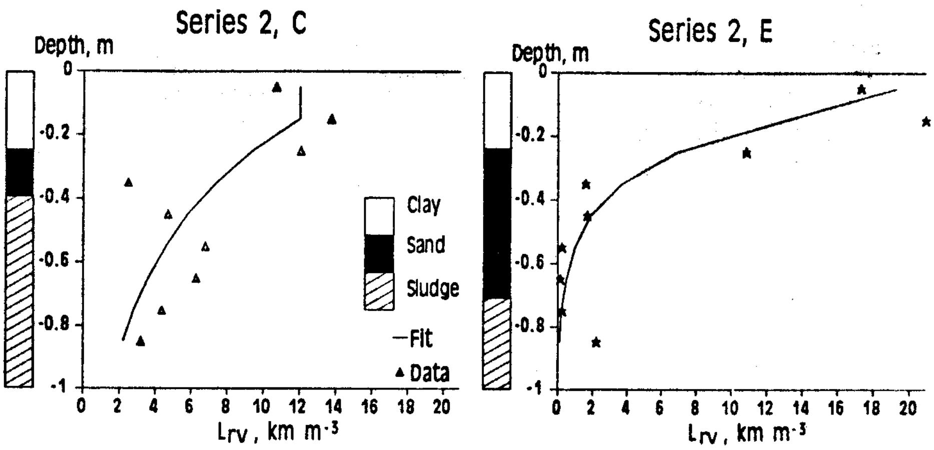


*Fig. 3.* As Figure 2, for the profiles with a sandy intermediate layer.

*Metal distribution in the profile*

The distribution of the total-metal concentrations in the profile (Table 4) shows no significant migration from the polluted subsoil to the unpolluted cover soil. The metal concentrations in the soil solution in the polluted and unpolluted soils differ much less than the total (hot-acid-extractable) concentrations (Table 4). The metal levels increase in the order: sand layer, cover layer and polluted soil, but the differences are not consistent. The conditions under which the soil was sampled were unfavourable: the soil was extremely wet because of heavy rainfall, and the presence of maize roots could give erratic results by releasing heavy metals during the extreme centrifugal forces during extraction.

The results suggest, however, that it is not very likely that increased metal levels in the plant result from migration of polluted soil solution into the unpolluted cover layer.

## Discussion

Before we discuss the results in the light of the questions raised in the introduction, we have to consider the reliability of the methods used. Although the $r^2$ values of the various regressions on root data are not very high (and do reflect the "quick and dirty" nature of core break methods), combining core break with limited calibrations seems to have been an appropriate choice for the current experiment.

From the two possible pathways for uptake of Cd from the river sediments, root penetration into the polluted layer was probably the most important. No evidence was obtained for substantial amounts of Cd or Zn moving upwards. Maize root distribution was not influenced by the depth at which the transition of topsoil to harbour sludge occurred. An intermediate sandy layer was only partially effective in preventing root development in the river sediment. The mini-plot experiment did not suffer from artificial root development along the edges into deeper layers, and the results may therefore be considered as valid indications for a field response, provided that the field heterogeneity, e.g. in applying a top layer, is taken into account.

A deeper groundwater table leads to a deeper root development and increases the required thickness of the clean top layer. No evidence was found for the concern that a shallower water table facilitates upward transport of metals into the top soil, at least not in the first ten years.

An exponential decrease of root length density with depth can be used as a rough first approximation of actual root distribution, but considerable deviations may occur (Table 3).

Although the difference in Cd and Zn uptake from the various profiles can be largely explained by the different relative amounts of roots in the polluted layer, the Cd uptake per unit root length was relatively high in the deeper layers, especially when we consider the probably shorter time available for uptake from deeper layers due to the time required for reaching this depth. The higher soil water content and thus increased mobility of metal ions apparently at least makes up for the shorter time available.

The experiment illustrates some of the problems in defining and quantitatively assessing "bioavailability" of pollutants: increasing the thickness of the top

*Table 4.* Heavy metals in soil and in soil solution

| Exp. series | Soil layer (m) | Polluted/ unpolluted | Cu Total (mg kg$^{-1}$) | Cu Solution ($\mu$g L$^{-1}$) | Zn Total (mg kg$^{-1}$) | Zn Solution ($\mu$g L$^{-1}$) | Cd Solution ($\mu$g L$^{-1}$) |
|---|---|---|---|---|---|---|---|
| 1,2 | 0–0.4 | Unpoll. | 13 | 46 | 97 | 59 | 2.34 |
| 1 | 0.6–0.8 | Unpoll. | 10 | 30 | 103 | 23 | 1.03 |
| 2 (sand) | 0.3–0.65 | Unpoll. | 3 | 25 | 11 | 9 | 0.41 |
| 1 | 0–0.4 | Polluted | 196 | 81 | 1256 | 147 | 3.26 |
| 1,2 | 0.6–0.8 | Polluted | 187 | 73 | 1276 | 112 | 3.38 |
| 3 | 0–0.4 | Unpoll. | 25 | 39 | 106 | 143 | 4.91 |
| 3 | 1.0–1.8 | Unpoll. | 24 | 28 | 104 | 69 | 3.08 |
| 3 | 0–0.4 | Polluted | 161 | 72 | 922 | 153 | 4.56 |
| 3 | 1.0–1.8 | Polluted | 153 | 66 | 973 | 244 | 6.16 |

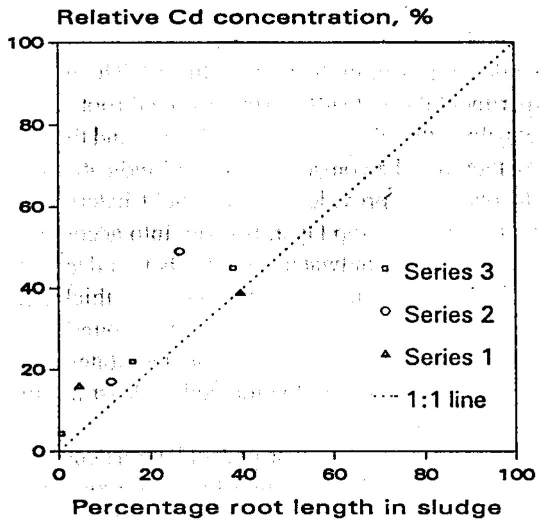


*Fig. 4.* Relative Cd content of maize as a function of the fraction of total root length in the harbour sludge for seven profiles; the Cd content of maize grown on pure harbour sludge was taken as 100% and the Cd content of maize grown on pure topsoil as 0%.

layer caused a gradual decrease in Cd uptake, roughly proportional to the relative root length in the Cd-containing zone. If soil sampling is to be used as basis for predicting Cd uptake by crops, the Cd distribution with depth within the complete rooted profile should be taken into account. Restriction to an "effective rooting depth" (as defined for soil surveys, McKeague et al., 1984) may underestimate uptake from deeper layers. A tentative index of Cd uptake might be constructed by weighing the concentration in each layer with the relative root length in that layer.

The absolute values of Cd uptake can not (yet) be explained in mechanistic terms, as a number of important processes are insufficiently known (Harmsen, 1992; Smilde et al., 1992). The data presented, however, indicate that reasonable predictions of Cd uptake in a new situation will be possible if we know the Cd uptake of plants grown on the clean top soil alone, plants grown on the polluted soil alone and if we know the relative root length in the polluted layer under actual field conditions. For future applications it may not be necessary to repeat experiments with large numbers of possible profile compositions. The interactions between root development and the water balance, however, should get careful consideration if one wants to cover up polluted soil layers as an alternative to expensive soil cleaning operations.

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