The Science of the Total Environment, 25 (1982) 225—244
Elsevier Scientific Publishing Company, Amsterdam — Printed in The Netherlands

CONSTRAINTS IN CROPPING HEAVY-METAL CONTAMINATED FLUVIAL SEDIMENTS

K.W. SMILDE, W. VAN DRIEL and B. VAN LUIT

Institute for Soil Fertility, P.O. Box 30003, 9750 RA HAREN (Gr.) (The Netherlands)

(Received May 11th, 1982; accepted June 16th, 1982)

ABSTRACT

Growth and heavy-metal uptake of various food crops and grass cultivated on harbour dredge spoils were studied, and health aspects in consuming the marketable products were discussed.

Vegetables (potato, carrot, radish, endive, lettuce) and grass (English ryegrass) performed well on dredge spoils, but small grains (wheat, barley) were affected by manganese deficiency. As compared with crops grown on uncontaminated reference soils, there was a net accumulation of As and heavy metals, especially so Cd, Zn and Cu, and a reduced uptake of Mn. Mainly because of the elevated Cd concentrations of the edible parts, exceeding the guideline of 0.1 mg/kg in fresh matter, the harbour dredge spoils investigated are considered unfit for the production of food crops, but may be used as grassland for dairy cattle. Highest Cd concentrations were attained in leafy vegetables and wheat (grain) and lowest in potato (tuber).

INTRODUCTION

There is a continuous transport of sediment into The Netherlands by the rivers Rhine and Meuse amounting to some 8×10^6 tons of wet material annually. The fluvial sediments are considerably contaminated by heavy metals resulting from upstream industrial activities. These sediments are deposited on river embankments (flood plains) and downstream in estuaries. There is also a flux of largely uncontaminated suspended material of marine origin into the estuaries. As a result, pollution of sediments deposited in river estuaries decreases downstream.

For the very large (crude oil) vessels to get access to the Rotterdam harbour basins, large-scale dredging operations are needed to remove deposited fluvial and marine sediments. Out of a total of some 20×10^6 cubic metres of dredged materials, annually, about 70% is of marine origin and is returned to the sea. This holds for the outer harbours adjacent to the sea. Dredge spoils of the inner harbours are much more severely polluted and have been deposited on polder land. This was done by pumping, which caused coarser and finer particles to segregate. Therefore, considerable differences in texture may occur over short distances. The finer particles contain most of the metal bonding adsorption sites. After a process of ripening, by drainage and leaching of excess salts by rain, the dredge spoils may be cropped. Studies on growth and heavy-metal uptake of various food crops and grass cultivated on dredge spoils are reported below, and health aspects are discussed.

MATERIALS AND METHODS

First experiment

Soils. Dredge spoils were selected on the basis of site of dredging (inner or outer harbour), texture and year of disposal. The first pot experiment comprised dredged materials from four disposal sites and with two textures ('light' and 'heavy'), and two reference (fluvial clay/loam) soils, with each substrate in six replicates (= six blocks). The terms 'light' and 'heavy' have a relative meaning here and only refer to differences in texture within each disposal site. Table 1 shows characteristics for representative dredge spoils (two disposal sites) and for the reference soils.

Crops. Carrot (cv. Amsterdamse bak, 0.04 g seeds per pot) and radish (cv. Cherry Belle, 30 seeds per pot) were seeded directly in 10-l, and spring wheat (cv. Selpek; 36 seeds per pot) and English ryegrass (0.3 g seeds per pot) in 6-l pots, whereas potato (cv. Eba) and butter head lettuce (cv. Deci Minor) were planted in 25- and 10-l pots, respectively, at three sprouts or seedlings per pot. Pots were placed in a glasshouse and each crop was treated as a separate experiment. Substrates were given sufficient N, P, K and Mg as reagent grade chemicals. Plants were watered at least once daily to 70% of field capacity with deionized water. Crops were harvested when mature, and samples were taken for analysis of N and several metals. For comparison, consumable parts from some field crops (potato, spring wheat, grass) growing on similar dredge spoils (Steendijkpolder, Broekpolder) were also sampled.

Analytical methods. Plant material was washed thoroughly in a 0.8% Teepol solution in deionized water and due care was taken to avoid metal contamination in the process of drying, chopping and grinding. Samples were wet-ashed with H_2SO_4/HNO_3 (for analysis of Cu, Mn, Ni, Zn) or HNO_3 (for Cd, Pb) and metal concentrations were determined by atomic absorption spectrophotometry. As and Hg were estimated by neutron activation analysis. N-Kjeldahl was determined colorimetrically in an autoanalyzer after digestion.

Soil samples for the selection of substrates were taken from the 0–20 cm top layer, dried at 35°C and crushed in a mill to pass a 2-mm sieve. Organic matter was estimated by oxidation with $K_2Cr_2O_7/H_2SO_4$ whereby excess $K_2Cr_2O_7$ is titrated. $CaCO_3$ was determined according to Scheibler and the fraction $< 16\,\mu m$ by the mass pipet method. Digestion and analytical procedures for heavy metals were similar for plant samples.

TABLE 1

CHEMICAL COMPOSITION, ON DRY MATTER BASIS, OF CONTAMINATED HARBOUR DREDGE SPOILS DEPOSITED IN POLDERS (1, 2) AND OF UNCONTAMINATED FLUVIAL CLAY/LOAM, REFERENCE, SOILS (3, 4) USED IN THE FIRST TWO EXPERIMENTS

Site/year of disposal	$CaCO_3$	Org. matter	Particles < 16μm	Fe	As	Cd	Cu	Hg	Mn	Ni	Pb	Zn
	g/100 g				mg/kg							
1a. Steendijkpolder 1*/1964	10.2	5.8	19.2	1.93	47	5.3	103	3.2	433	28	214	790
1b. Steendijkpolder h/1964	13.5	10.5	37.3	3.63	108	10.8	233	8.4	728	54	424	1660
2a. Kralingerpolder 1/1972	12.3	8.8	33.0	2.65	35	8.9	113	4.5	631	39	180	697
2b. Kralingerpolder h/1972	12.0	13.8	52.5	3.79	69	16.3	206	9.2	790	66	331	1240
3. Fluvial loam Kesteren	10.6	1.3	26.2	1.87	8	0.2	12	0.04	659	25	19	58
4. Fluvial clay Nieuwveen	2.0	5.2	55.2	2.94	15	0.3	14	0.07	511	32	29	83

*1 and h denote 'light' and 'heavy' substrates, respectively within each disposal site.

Second experiment

In a separate experiment, oats (cv. Leander) and spring wheat (cv. Kaspar) were grown on the substrates used in the first experiment to study the incidence and control of Mn-deficiency. Substrates were homogenized and sieved to remove root residues. An additional N, P, K, Mg dressing was supplied. When Mn-deficiency symptoms ('grey speck') occurred, six of the twelve replicates per treatment (= substrate) received a foliar application of a solution containing 12.5 g $MnSO_4 \cdot H_2O$ and 2.5 g hydrated lime per litre. The foliar spray was repeated some three weeks later.

Third experiment

In this experiment crops were grown on mixtures of uncontaminated fluvial clay, reference, soil from the Noordbovenpolder and dredge spoils from the Oostabtspolder. The mixtures contained 0, 3, 6, 10, 13, 17, 20, 24, 27 or 31 percent (by volume) dredge spoils, respectively. Cd, Cu, Pb and Zn concentrations of the mixtures increased linearly with the percentage dredge spoils. By contrast, Mn and Ni concentrations were similar in the reference soil, the dredge spoils and the various mixtures. Some pertinent data are presented in Table 2. Each treatment (= substrate) was replicated 4 times (4 blocks). Potato (cv. Eba), carrot (cv. Amsterdamse Bak), butter head lettuce (cv. Reskia), spring wheat (cv. Melchior) and English ryegrass were grown under conditions similar to those of the first experiment. After harvest, substrates were homogenized and sieved, and additional N, P, K and Mg was applied prior to sowing or planting potato (cv. Eba), radish (cv. Cherry Belle), endive, spring barley (cv. Aramir) and English ryegrass. Cereal crops were sprayed with manganese sulphate to control Mn-deficiency.

RESULTS AND DISCUSSION

Crop yield

(a) Dredge spoils versus fluvial clay/loam. Crops performed well on dredge spoils as compared with fluvial clay/loam, reference, soils. Table 3 shows representative data for the first experiment. In general, yields were higher on the finer textured substrates. Spring wheat grown on dredge spoils is prone to Mn deficiency, in some cases resulting in severe yield reduction: no close relationship between substrate-Mn and incidence of Mn deficiency was found. More comprehensive information is given by Van Driel et al. (1977).

In the second experiment, spring wheat and oats showed slight to severe symptoms of Mn-deficiency when grown on dredge spoils. Depending on the substrate, one or two foliar sprays of manganese sulphate were required to control the disorder. This treatment increased both grain and straw yield of the two crops (highly) significantly. Responses differed among substrates but were not found to be related to texture and chemical compo-

TABLE 2

CHEMICAL COMPOSITION, ON DRY MATTER BASIS, OF UNCONTAMINATED FLUVIAL CLAY, REFERENCE, SOIL (NOORDBOVENPOLDER), CONTAMINATED DREDGE SPOILS (OOSTABTSPOLDER), AND MIXTURES OF THESE SUBSTRATES, AS USED IN THE FIRST EXPERIMENT.

Substrate	$CaCO_3$	Org. matter	Particles < 16μm	pH-KCl	Cd	Cu	Mn	Ni	Pb	Zn
	g/100 g				mg/kg					
a. Noordbovenpolder	7.3	6.2	65.1	7.1	0.50	31	1150	50	46	138
b. Oostabtspolder	14.7	13.4	57.2	7.1	18.0	183	902	62	255	1119
90% a + 10% b	7.7	6.8	63.8	7.2	1.98	45	1060	48	67	220
80% a + 20% b	8.6	7.6	62.3	7.2	3.46	58	1090	50	84	296
69% a + 31% b	8.8	8.0	62.4	7.2	5.09	74	1040	54	100	407

TABLE 3

DRY MATTER YIELDS (in g per pot) OF EDIBLE PARTS OF VARIOUS CROPS GROWN ON CONTAMINATED DREDGE SPOILS (1, 2) OR FLUVIAL CLAY/LOAM, REFERENCE, SOILS (3, 4)
Results of the first experiment; means of 6 replicates.

Site of disposal	Crop					
	Potato (tuber)	Carrot (root)	Radish (root)	Lettuce (head)	Spring wheat (grain)	Grass* (leaf)
1a. Steendijkpolder 1**	870	61	7.2	7.7	11.9	34
1b. Steendijkpolder h	970	63	6.2	9.8	34.9	39
2a. Kralingerpolder 1	1090	58	4.0	7.4	21.4	40
2b. Kralingerpolder h	1250	63	4.0	10.0	28.2	44
3. Fluvial loam Kesteren	620	56	7.0	7.9	32.6	37
4. Fluvial clay Nieuwveen	960	62	5.2	10.0	32.1	42

*Sum of 3 cuts.
**1 and h denote 'light' and 'heavy' substrates, respectively, within each disposal site.

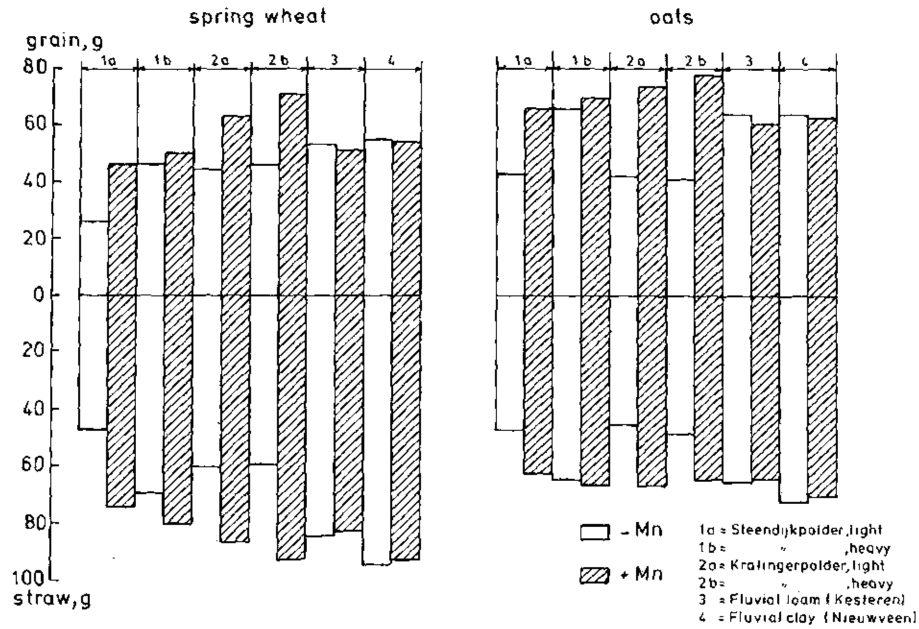

Fig. 1. Grain and straw dry matter yields (g per pot) of cereal crops (left spring wheat; right oats) as affected by foliar Mn application when grown on contaminated dredge spoils (1, 2) or fluvial clay/loam, reference, soils (3, 4). Results of the second experiment; means of 6 replications.

sition. The fluvial clay/loam, reference, soils supported crops that neither showed Mn deficiency nor responded to applied Mn. Some pertinent data are illustrated in Fig. 1. For more detail reference is made to Smilde and Van Driel (1977).

(b) Mixtures of dredge spoils and fluvial clay. Most crops responded positively to substituting dredge spoils for fluvial clay, at least up to 10–20 percent by volume; higher doses tended to depress yields. Representative data are depicted in Table 4. For more detailed results see Smilde and Van Driel (1979).

Crop chemical composition

(a) Dredge spoils versus fluvial clay/loam. Chemical data on the edible parts of the various food crops and on grass are presented in Tables 5–7 (first experiment). Crops grown on contaminated dredge spoils contained more As and heavy metals than did crops on uncontaminated fluvial clay/loam soils used as a reference. This is true both for edible parts and foliage. The data for foliage, when not edible, are not shown here. Concentrations were highest in the foliage, but in the case of Cu in (wheat) grain. Accumulation of As and heavy metals by crops cultivated on contaminated dredge spoils is clearly demonstrated by the 'accumulation factors' for the various elements, i.e. the mean concentration in the crops grown on the dredge spoils over the mean concentration in the crops on the reference soils.

TABLE 4

DRY MATTER YIELDS (in g per pot) OF EDIBLE PARTS OF VARIOUS CROPS GROWN ON FLUVIAL CLAY, REFERENCE, SOIL (NOORDBOVENPOLDER), OR MIXTURES OF THIS SOIL WITH CONTAMINATED DREDGE SPOILS (OOSTABTSPOLDER)
Results of the third experiment; means of 4 replicates.

Substrate	Potato (tuber)		Carrot (root)	Radish (root)	Lettuce (head)	Endive (leaf)	Spring wheat (grain)	Spring barley (grain)	Grass (leaf)*	
	(1)**	(2)	(1)	(2)	(1)	(2)	(1)	(2)	(1)	(2)
100% clay	300	170	40	6.9	17	13.8	31	44	38	66
90% clay + 10% dredge spoils	330	170	45	6.4	17	12.1	38	57	76	71
80% clay + 20% dredge spoils	340	230	44	6.0	19	12.2	33	55	75	73
69% clay + 31% dredge spoils	330	280	48	6.0	19	11.4	33	59	64	67

*Sum of 4 cuts.
**1 = first; 2 = second growing season.

TABLE 5

CHEMICAL COMPOSITION OF POTATO TUBER AND CARROT ROOT, in mg/kg dry matter (Hg in μg/kg), ON CONTAMINATED DREDGE SPOILS (1, 2) AND FLUVIAL CLAY/LOAM, REFERENCE, SOILS (3, 4)

Results of the first experiment. For conversion to fresh weight basis multiply by 0.27 or 0.108, for potato tuber and carrot root, respectively. Concentrations exceeding tentative guidelines for healthy foodstuffs are underlined.

Site of disposal	Potato tuber								Carrot root							
	As	Cd	Cu	Hg	Mn	Ni	Pb	Zn	As	Cd	Cu	Hg	Mn	Ni	Pb	Zn
1a. Steendijkpolder 1*	0.15	0.17	18.4	21	2.0	0.58	0.96	18	0.33	0.60	8.9	30	5.2	0.37	0.74	31
1b. Steendijkpolder h	0.13	0.17	8.7	16	3.1	0.17	0.41	14	0.27	0.63	8.8	25	6.0	0.34	0.66	32
2a. Kralingerpolder 1	0.07	0.21	8.3	13	3.1	0.13	0.41	16	0.17	1.05	8.7	21	6.7	0.52	0.53	34
2b. Kralingerpolder h	0.08	0.21	11.1	12	3.1	0.24	0.33	18	0.21	1.65	10.4	23	6.2	0.65	0.52	37
3. Fluvial loam Kesteren	0.02	0.08	5.1	5.3	2.4	0.13	0.11	10	0.04	0.12	5.6	8.5	10.0	0.23	0.15	14
4. Fluvial clay Nieuwveen	0.02	0.08	4.6	4.9	2.0	0.07	0.11	10	0.11	0.13	4.3	7.9	5.1	0.31	0.12	14
Accum. factor**	2.8	2.0	2.2	1.2	0.4	1.1	1.1	2.0	2.4	4.1	1.6	1.6	0.5	1.4	1.5	2.5

*1 and h denote 'light' and 'heavy' substrates, respectively, within each disposal site.

**ratio mean foliar concn. in crops grown on dredge spoils/mean foliar concn. in crops grown on reference soils.

TABLE 6

CHEMICAL COMPOSITION OF RADISH ROOT AND LETTUCE HEAD, In mg/kg dry matter (Hg in μg/kg), ON CONTAMINATED DREDGE SPOILS (1, 2) AND FLUVIAL CLAY/LOAM, REFERENCE, SOILS (3, 4)

Results of the first experiment. For conversion to fresh weight basis multiply by 0.049 or 0.076 for radish and lettuce, respectively. Concentrations exceeding tentative guidelines for healthy foodstuffs are underlined.

Site of disposal	Radish root								Lettuce head							
	As	Cd	Cu	Hg	Mn	Ni	Pb	Zn	As	Cd	Cu	Hg	Mn	Ni	Pb	Zn
1a. Steendijkpolder 1*	1.91	0.7	9	84	10.0	0.9	3.9	83	0.50	2.41	9.5	48	21	1.39	0.71	95
1b. Steendijkpolder h	2.09	0.9	9	95	10.0	0.7	4.4	90	0.45	2.80	10.7	39	26	1.25	0.76	107
2a. Kralingerpolder 1	0.84	0.8	8	47	15.0	0.7	1.8	66	0.28	3.46	12.7	33	18	2.24	0.76	119
2b. Kralingerpolder h	1.31	1.7	11	71	10.1	1.3	2.7	126	0.27	6.48	12.8	28	23	1.48	0.56	134
3. Fluvial loam Kesteren	0.31	0.3	5	12	18.0	0.9	0.8	42	0.07	1.18	8.6	25	44	2.00	0.76	58
4. Fluvial clay Nieuwveen	0.38	0.2	5	8.2	14.0	0.6	0.5	30	0.32	1.01	5.7	26	38	1.09	0.36	32
Accum. factor**	1.7	4.2	1.6	1.3	0.6	1.3	1.8	3.1	1.6	3.4	1.7	1.3	0.5	0.8	1.1	2.5

*1 and h denote 'light' and 'heavy' substrates, respectively, within each disposal site.

**ratio mean foliar concn. in crops grown on dredge spoils/mean foliar concn. in crops grown on reference soils.

TABLE 7

CHEMICAL COMPOSITION OF SPRING WHEAT GRAIN AND GRASS FOLIAGE (SECOND CUT), in mg/kg dry matter (Hg in μg/kg), ON CONTAMINATED DREDGE SPOILS (1, 2) AND FLUVIAL CLAY/LOAM, REFERENCE, SOILS (3, 4)

Results of the first experiment. For conversion to fresh weight basis multiply by 0.90 for spring wheat. Concentrations exceeding guidelines for healthy foodstuffs or feedstuffs are underlined.

Site of disposal	Spring wheat grain								Grass foliage							
	As	Cd	Cu	Hg	Mn	Ni	Pb	Zn	As	Cd	Cu	Hg	Mn	Ni	Pb	Zn
1. Steendijkpolder 1*	0.09	0.29	9.7	1.0	1.9	0.52	0.71	87	0.77	0.29	18.0	21	12.3	0.92	0.63	188
1b. Steendijkpolder h	0.06	0.48	8.2	< 1.0	8.2	0.30	0.60	57	0.45	0.36	20.4	17	35.8	1.81	0.74	204
2a. Kralingerpolder 1	0.04	0.78	10.5	< 1.0	3.8	0.39	1.39	113	0.34	0.45	19.4	21	17.2	1.34	1.08	152
2b. Kralingerpolder h	0.04	0.84	11.4	< 1.0	6.9	0.30	0.79	111	0.42	0.59	23.4	19	23.2	2.17	0.86	194
3. Fluvial loam Kesteren	0.01	0.07	4.4	< 1.0	21.0	0.18	0.71	22	0.09	0.10	9.7	14	45.5	0.73	0.77	46
4. Fluvial clay Nieuwveen	0.03	0.06	3.5	< 1.0	26.0	0.01	0.71	24	0.29	0.09	7.0	19	35.4	0.94	0.53	49
Accum. factor**)	3.0	6.0	6.6	2.0	0.4	1.4	2.0	15.1	2.4	4.7	2.5	1.4	0.4	1.7	1.5	3.6

*1 and h denote 'light' and 'heavy' substrates, respectively, within each disposal site.
**ratio mean foliar concn. in crops grown on dredge spoils/mean foliar concn. in crops grown on reference soils.

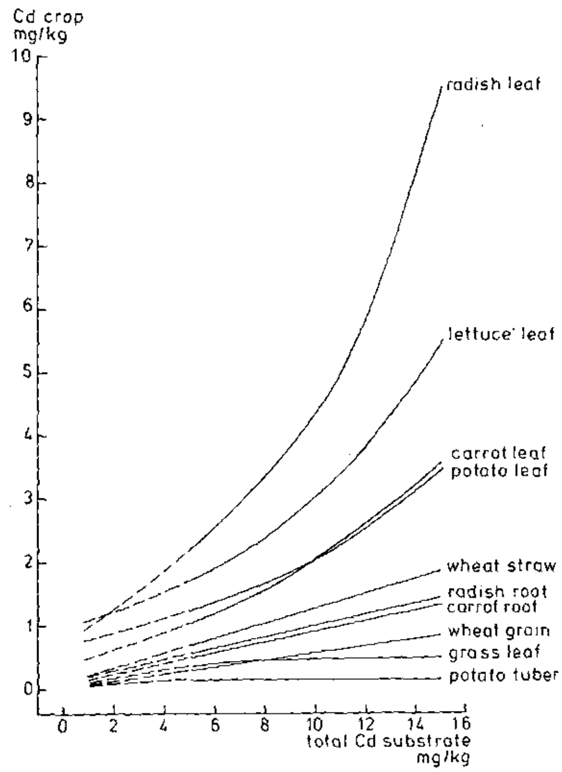

Fig. 2. Relationship between total Cd in the substrate (dredge spoils and fluvial clay/loam) and in the plant.

Tables 5–7 present accumulation factors for plant foliage. Accumulation factors, or foliar concentration ratios, varied among elements, descending in the order Cd, Zn > Cu, As > Hg, Pb, Ni. Weak accumulation of Ni is associated with the low Ni content of dredged materials as compared with clay/loam, reference, soils.

Plants accumulated less Mn on the dredge spoils than on the reference soils, as shown by the accumulation factors of less than 1. Impeded Mn uptake from dredge spoils is associated with high organic matter contents combined with high calcium carbonate, and also with high Zn. In cereal crops this may induce Mn deficiency, or a Mn deficiency/Zn toxic syndrome, judging from the high Zn concentration of wheat.

The higher As and heavy-metal levels in the finer textured substrates were not reflected in the plant's chemical composition, except for Cd and Zn, as shown in Figs. 2 and 3. The data comprise all dredge spoils investigated, i.e. four disposal sites and two textures (p. 226). Dotted parts of the curves are speculative because of the large gap in Cd/Zn concentrations between dredge spoils and clay/loam, reference, soils. The relationship between plant and substrate Cd/Zn concentrations is best established for plant foliage.

Contrary to expectations, there is no evidence that ripening of the dredge

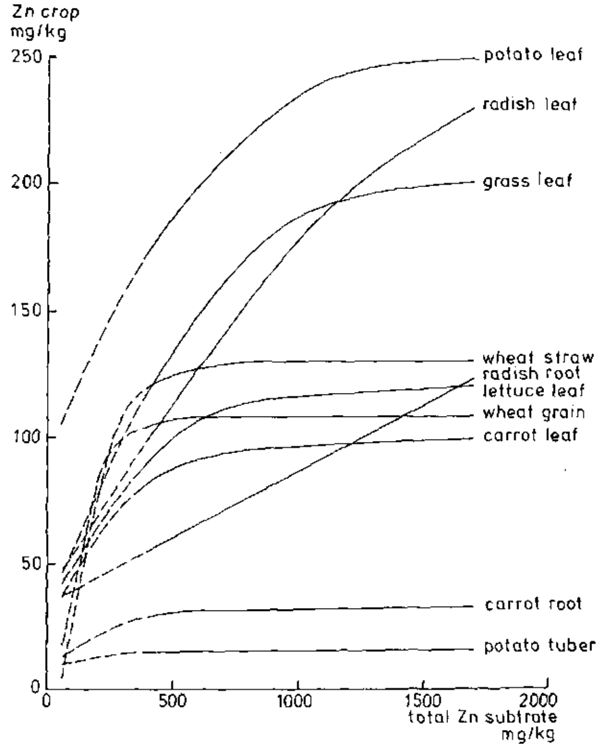

Fig. 3. Relationship between total Zn in the substrate (dredge spoils and fluvial clay (loam) and in the plant.

spoils, characterized by organic matter decomposition and calcium carbonate losses, favoured element mobility resulting in increased plant availability. As for Cd, crops tended to accumulate even more of this metal on the younger, unripened dredge spoils (Kralingerpolder) than on the more ripened ones (Steendijkpolder).

The effect of treating cereal crops with a manganese sulphate spray on plant chemical composition (second experiment) is shown in Fig. 4 for wheat grain. Trends for straw are similar, both in wheat and oats, but Mn concentrations in straw are unduly high as spray residues cannot be fully removed before plant analysis. Manganese application considerably raised Mn and lowered other heavy metal and As concentrations of the crop grown on dredge spoils. By contrast, plant chemical composition on the clay/loam, reference, soils was little affected by treatment with Mn, apart from a slight rise in grain—Mn and a substantial (apparent) increase in straw Mn. These findings are largely explained by dilution, compare Figs. 1 and 4. Actually, there is a marked growth response and a concomitant decrease in element concentrations other than Mn in the Mn-deficient crop on the dredge spoils following Mn-treatment. By contrast, there is no growth response and no change in chemical composition, apart from Mn, in the Mn-sufficient crop

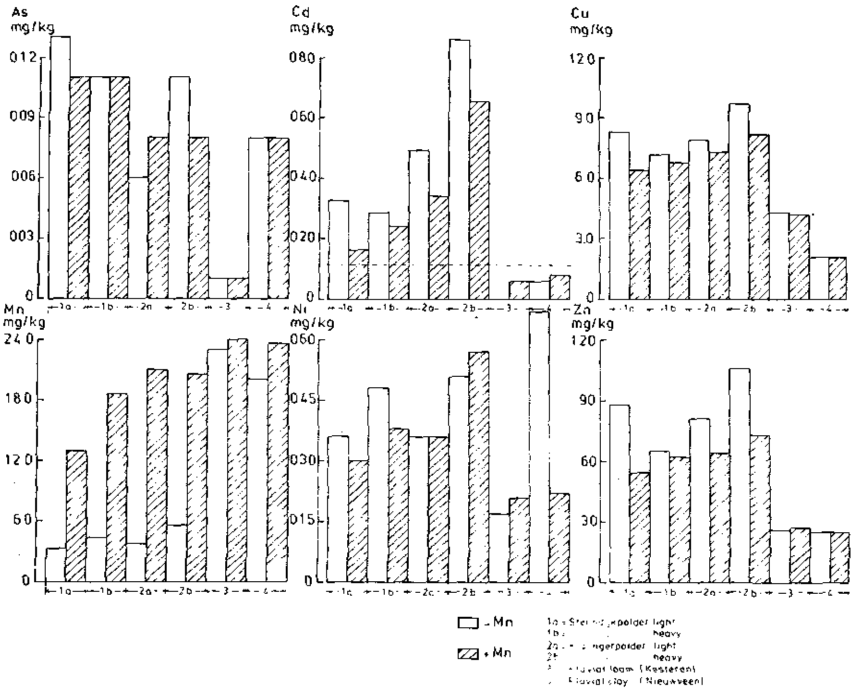

Fig. 4. Chemical composition of wheat grain (in mg/kg dry matter), as affected by foliar Mn application when grown on contaminated dredge spoils (1, 2) or fluvial clay/loam, references, soils (3, 4). Results of the second experiment. ———— = tentative guideline for maximum permissible Cd concentration in cereal grain.

on the fluvial clay/loam soils upon Mn-treatment. No explanation can be offered for the erratic Ni-concentration in the grain of the untreated wheat crop on fluvial clay.

The Mn-status of the cereal crops on dredged spoils is typical of Mn-deficiency (Coïc and Coppenet, 1958). However, as pointed out before, Zn-toxicity cannot be excluded as a causative factor. In fact, Zn-induced Mn-deficiency is a well-known phenomenon, for instance in sugar beet (Hewitt, 1953) and oats (Smilde, 1976).

(b) Mixtures of dredge spoils and fluvial clay. Subsititution of dredge spoils for fluvial clay (third experiment) raised heavy-metal concentrations in the plant, but there was a pronounced difference among elements. Plant Cd and Zn were positively related to (total) substrate contents, the levels increasing with increasing proportions dredge spoils in the substrate (Figs. 5 and 7). This also holds for Cu in some plant species, such as grass and leafy vegetables, but not in others (Fig. 6). There is little congruency between plant and substrate (total) Pb; the data are not represented here. As in the first experiment (Figs. 2 and 3) plant foliage was generally higher in heavy metals than were the other plant parts. For the sake of clearness Figs. 5–7 only depict data for edible parts. On the basis of the evidence presented so

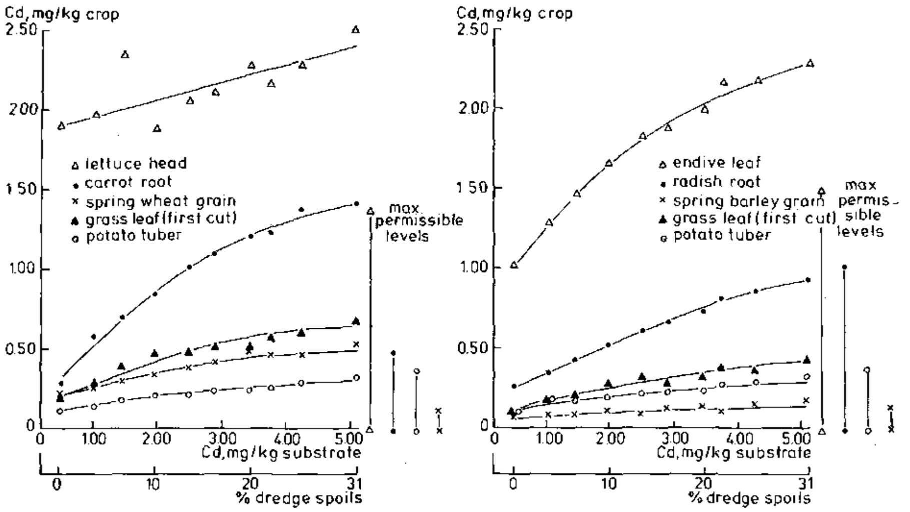

Fig. 5. Cd concentrations (mg/kg) dry matter) of edible parts of various crops grown on fluvial clay (Noordbovenpolder) or mixtures of this soil with dredge spoils (Oostabtspolder). Results of third experiment; left first, right second growing season.

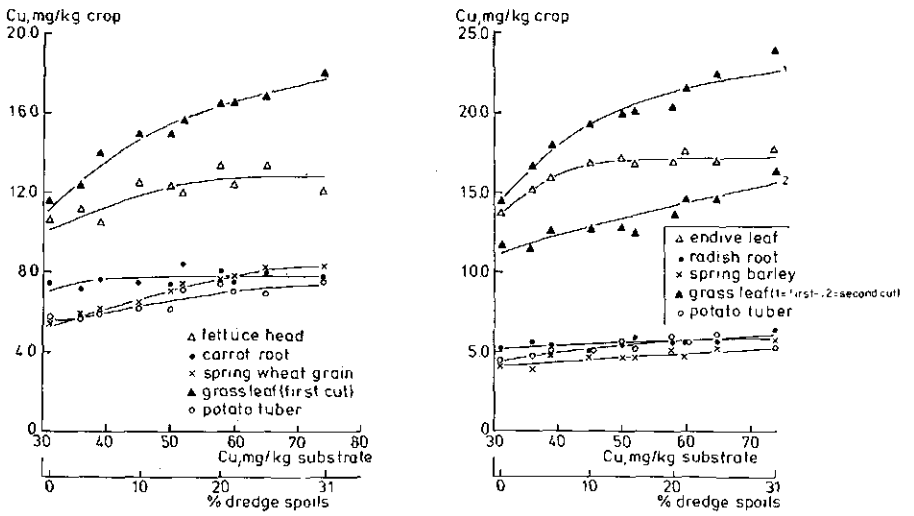

Fig. 6 Cu concentrations (mg/kg dry matter) of edible parts of various crops on fluvial clay (Noordbovenpolder) or mixtures of this soil with dredge spoils (Oostabtspolder). Results of third experiment; left first, right second growing season.

far it is postulated that leafy vegetables show a preference for Cd, Cu and Zn, and grass for Cu and Zn accumulation, whereas potato tubers accumulate only small amounts of these metals.

(c) Dredge spoils; commercially grown crops. Glasshouse and simultaneously grown field crops in the Broekpolder and Steendijkpolder, i.e.

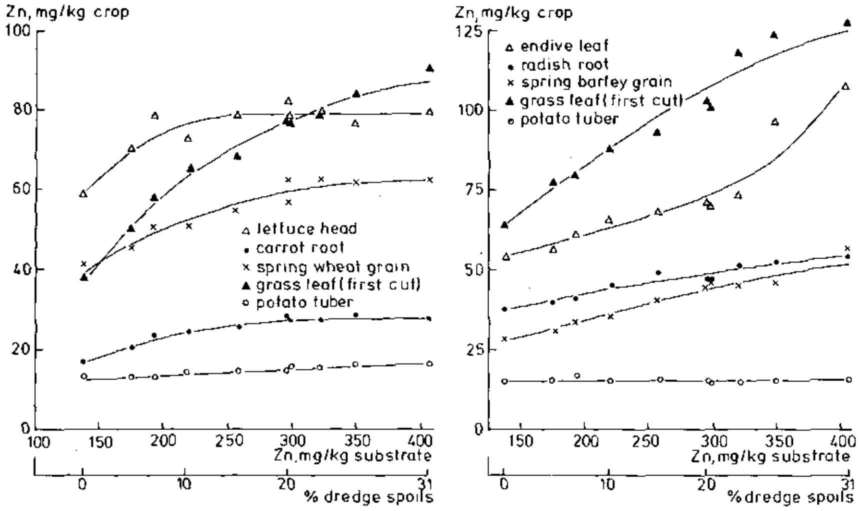

Fig. 7. Zn concentrations (mg/kg dry matter) of edible parts of various crops grown on fluvial clay (Noordbovenpolder) or mixtures of this soil with dredge spoils (Oostabtspolder). Results of third experiment; left first, right second growing season.

potatoes, spring wheat and grass, showed no consistent differences in chemical composition. However, there is a notable exception for the much higher Pb content of grass grown in the open. This may be due to atmospheric pollution, which has a greater impact on exposed plant parts than on grain and tuber. No obvious explanation can be offered for the higher Cd concentrations in wheat grain and grass, and the Zn and Cu concentrations in wheat grain and potato tuber, of the field crops. The data are not presented here.

Health aspects

Standards have been set by WHO for heavy-metal contaminants considered hazardous to human health, the so-called acceptable daily intake (ADI), amounting to 65, 45 and 430 μg for Cd, Hg and Pb, respectively (WHO, 1972). A tentative limit of 3000 μg per day has been suggested by WHO for As (WHO, 1973). Based on ADI, and taking into account the variation in native heavy-metal concentrations of the diet constituents, and the composition of the diet, guidelines for maximum heavy-metal concentrations in the foodstuffs composing the diet may be given.

Tentative maxima for Cd, Hg and Pb in foodstuffs, as proposed by the German Ministry of Health (An., 1979), are shown in Table 8. In the Netherlands daily intake of Cd and Pb so far remains well below WHO/ADI standards (Min. Volksgezondheid Milieuhygiëne, 1980); no data on Hg are provided. When using the German guidelines for Cd and Pb in foodstuffs in the consumption pattern common in the Netherlands (LEI, 1980), the ADI for these elements as proposed by WHO is slightly exceeded (Table 8). So German guidelines may have to be adapted to local conditions and consumption patterns.

TABLE 8

DAILY CONSUMPTION OF FOODSTUFFS, GUIDELINES FOR MAXIMUM Cd, Hg AND Pb CONCENTRATIONS (in μg/g = mg/kg fresh wt.) in foodstuffs, and present and acceptable daily intake of heavy metals.
In parentheses daily intake (ADI) according to WHO standards.

	Main human foodstuffs									
	Bread cereals	Vegetables		Potatoes	Fruits	Milk	Meat	Miscell.	Total daily intake (μg)
		leafy	root						
Daily consumption (g)*	155	200		210	130	440	160		
Cd									
present daily intake (μg)**	4.7	2.4		5.0	0.8	0.8	6.5	3.5	23.7
max. acceptable concn. (μg/g)***	0.1	0.1	0.05	0.1	0.05	0.0025	0.1		
max. accpetable daily intake (μg)	15.5	15.0	2.5	21.0	6.5	1.1	16.0		77.6 (65)
Hg									
present daily intake (μg)									
max. accpetable concn. (μg/g)	0.03	0.03	0.03	0.02	0.03	–	0.035		
max. acceptable daily intake (μg)	4.6	4.5	1.5	4.2	3.9	–	5.6		24.5 (45)
Pb									
present daily intake (μg)	22.4	15.7		6.6	8.0	4.2	7.9	26.9	91.7
max. acceptable concn. (μg/g)	0.5	1.2	0.5	0.2	0.5	0.05	0.3		
max. accpetable daily intake (μg)	77.5	180	25.0	42.0	65.5	22.0	48.0		460 (430)

*Landb. Econ. Inst./Centr. Bureau Statist., 1978 (see references).
**Min. van Volksgeozondh./Milieuhygiëne (Ministry of Health and Environment), 1980 (see references).
***B. Gesundheits B1. 22 (1979) Dtsch. Lebensm. Rundsch. 75 (1979), see references.

TABLE 9
MAXIMUM PERMISSIBLE CONCENTRATIONS OF SOME ELEMENTS IN FEEDS, (in mg per kg material with 12% moisture) AS ESTABLISHED BY THE EUROPEAN COMMUNITY (1974)

	Single feed	Mixed feed	Roughage
As	2	2	4
Cu		20*	20*
Hg	0.1	0.1	
Pb	10	5	40

*Guidelines used by the Dutch Ministry of Agriculture and Fisheries (1973).

Cd concentrations (expressed on fresh weight) of carrot root (Table 5), lettuce head (Table 6) and wheat grain (Table 7) grown on pure dredge spoils exceeded the tentative maxima in food, i.e. 0.05 mg/kg Cd in root vegetables, and 0.1 mg/kg in leafy vegetables and cereal grain (Table 8). This is also true for radish root established on dredge spoils containing well over 10 mg/kg total Cd (Table 6). Cd in potato tuber remained well below the limit of 0.1 mg/kg, even on dredge spoils with 16 mg/kg total Cd (Table 5). Spraying cereal crops on dredge spoils with manganese sulphate, though controlling Mn-deficiency and vigorously stimulating growth, did not reduce grain Cd to a level considered permissible (Fig. 4).

Pb concentrations of the edible parts of the various crops met the standards for healthy foodstuffs. However, in wheat grain the acceptable limit of 0.5 mg/kg (Table 8) was exceeded, not only in the crop grown on dredge spoils but also in that on the uncontaminated fluvial clay. It is difficult to offer a ready explanation for the high uptake of Pb from the fluvial clay with its 'normal' Pb content. Hg concentrations of all foodstuffs on dredge spoils remained well below the permissible levels for this element as stated in Table 8.

Both on the fluvial clay reference soil, containing 0.50 mg/kg total Cd, and the mixtures of this soil with dredge spoils, lettuce leaf and wheat grain Cd concentrations were found to surpass the acceptable maxima. Carrot root, endive leaf and barley grain attained the respective permissible limits when substituting 3, 6, and 10% dredge spoils for fluvial clay, respectively, the substrates containing 1.03, 1.46 and 1.98 mg/kg total Cd, respectively. Cd concentrations of potato tuber and radish root remained below the acceptable maxima, even on substrates containing 31% dredge spoils and 5.9 mg/kg total Cd (Fig. 5). Pb concentrations of the edible parts of all crops, including wheat, remained below the acceptable maxima; the data are not shown here.

Guidelines for maximum concentrations of As, Cu, Hg and Pb in feeds have been established by the European Community (1974), see Table 9. As shown in Table 7, As, Hg and Pb concentrations in grass grown on pure dredge spoils cannot be considered hazardous for animal health. However, grass Cu attained or exceeded the permissible limit of 20 mg/kg dry matter in sheep roughage (Min. Agriculture Fisheries, 1973). The substrates in this case contained 100 mg/kg total Cu or more. Cu concentrations of grass cultivated in mixtures of fluvial clay and dredge spoils, with up to 74 mg/kg total Cu, were below the acceptable maximum of 20 mg/kg, except in the first cut of the second growing season in the case of substrates containing 50 mg/kg total Cu or more (Fig. 6).

No guidelines for Cd in feeds are available yet. In a review Declaire and De Cat (1979) mention 0.6 mg Cd/kg dry matter for sheep as a tentative maximum, whereas acceptable levels for ruminants may be much higher. In the Netherlands for (dairy) cattle tentative levels of 0.5, 1.0 and 1.0–2.0 mg Cd/kg dry matter are suggested for single feed, mixed feed and roughage, respectively. In the present study the tentative maximum for sheep is attained in grass grown on fluvial clay enriched with dredge spoils, at least in some cuts (Fig. 5) but, surprisingly, not in grass grown on pure dredge spoils (Table 7).

From the above results it is concluded that the use of contaminated dredge spoils for cultivation of food crops, with a possible exception for potatoes, should be discouraged because of the heavy-metal (mainly Cd) enrichment of the edible parts. Dredge spoils may be used as grassland for dairy cattle but not for sheep, mainly because of the high Cu content of the herbage.

CONCLUSION

Potato (*Solanum tuberosum* L.), carrot (*Daucus carota* L.), radish (*Raphanus sativus* L.), butter head lettuce (*Lactuca sativa* L.), endive (*Cichorium endiva* L.), spring wheat (*Triticum aestivum* L.), spring barley (*Hordeum vulgare* L.) and English ryegrass (*Lolium perenne* L.) were grown in pots on contaminated harbour dredge spoils, uncontaminated fluvial clay/loam (reference) soils, or mixtures of dredge spoils and fluvial clay. Crops performed well on dredge spoils, and on mixtures of dredge spoils and fluvial clay, but cereals were affected by manganese deficiency.

Crops grown on contaminated dredge spoils contained more Cd, Zn, Cu, As, Hg, Pb and Ni, and less Mn in leaf tissues and edible parts than did crops on the reference soils. There was no clear relationship between substrate and plant (foliar) metal concentrations, with an exception for Cd and Zn.

Crops established on mixtures of dredge spoils and fluvial clay accumulated, mainly in the foliage, Cd, Zn and to some extent Cu, according to the increase in the dredge spoils component of the substrate. There was little congruency between plant and substrate (total) Pb.

Cd concentrations of edible parts considered hazardous in human consumption were attained in wheat and lettuce when grown on fluvial clay containing 0.5 mg/kg (total) Cd, in carrot, endive and barley on fluvial clay/dredge spoils mixtures with Cd contents of 1.09, 1.46 and 1.98 mg/kg, respectively, in radish on pure spoils with at least 10 mg/kg Cd, but in potato not even on pure dredge spoils with 16 mg/kg Cd.

It is concluded that contaminated dredge spoils are unfit for cultivation of food crops, with a possible exception for potatoes, because of the Cd enrichment of the edible parts. Dredge spoils may be used as grassland for dairy cattle, but not for sheep, due to the high Cu and, sometimes, Cd contents in the herbage.

REFERENCES

Anonymous, Richwerte für Blei, Cadmium und Quecksilber in und auf Lebensmitteln. B. Gesundheits B1., 22 (1979) 282. Dtsch. Lebensm. Rundsch., 75 (10) (1979) 319.

Coïc, Y et M. Coppnent, Sur la carence en manganèse des céréales dans les sols humifères de Bretagne. Ann. Agron. Physiol. Végét. Suppl., II (1958) 111–38.

Declaire, M. en W. de Cat, Bibliografisch overzicht betreffende de gevoeligheid van huisdieren tegenover verontreinigd voedsel door koper, cadmium en lood. Landbouw Tijdschrift, 32 (1979) 1192–1198.

Driel, W. van, B.J. van Goor en K.W. Smilde, Zware metalen in havenslib en enkele daarop verbouwde gewassen. Inst. Bodemvruchtbaarheid, Nota 42, 1977, 21 pp.

European Community, Richtlijn Raad EEG (17/12/73) tot vaststelling van de maximale gehalten aan ongewenste stoffen en produkten in diervoeders. Publ. blad EEG. 17, 1974. L38: 31–36.

Hewitt, E.J., metal interrelationships in plant nutrition. I. Effects of some metal toxicities on sugar beet, tomato, oat, potato and marrowstem kale in sand culture. J. Exp. Bot., 4 (1953) 54–64.

Landbouw Economisch Instituut/Centraal Bureau voor de Statistiek, Landbouwcijfers, Den Haag 1980.

Ministerie van Volksgezondheid en Milieuhygiëne/Ministry of Health and Environmental Hygiene, Bewakingsprogramma 'Mens en Voeding', 1980, Rapp. 8.

Ministry of Agriculture and Fisheries, Committee on Mineral Nutrition, The Hague, Tracing and treating mineral disorders in dairy cattle. Centre for Agricultural Publishing and Documentation, Wageningen, 1973.

Smilde, K.W., Minor elements in the nutrition of cereals. Compte-Rendu Semaine d'Etude Cereal Crops, Gembloux, 1976, pp. 303–312.

Smilde, K.W. en W. van Driel, Mangaangebrek in granen verbouwd op havenslib. Inst. Bodemvruchtbaarheid, Nota 39, 1977, 19 pp.

Smilde, K.W. en W. van Driel, Een onderzoek naar het cadmium–, zink–, koper– en loodgehalte van het gewas in afhankelijkheid van de gehalten in de grond. Inst. Bodemvruchtbaarheid, Nota 67, 1979, 23 pp.

World Health Organization, Evaluation of certain food additives and the contaminants mercury, lead and cadmium. Techn. Rept. Series 505, 1972.

World Health Organization, Trace elements in human nutrition. Tech. Rept. Series 523, 1973.