WATER RESEARCH

WRC

741/1

Project Leader: R D Davis

Authors: C H Carlton-Smith, J H Stark, B A Thomas* and R D Post*

December 1986

DoE 1182-M/2

EFFECTS OF METALS IN SLUDGE ON CROPS (EI 9324 SLD)

Final Report to the Department of Environment

DoE Contract Reference: DGR/480/457
Contract Duration: November 1978 - March 1986

* Thames Water

WRc Environment,
Medmenham Laboratory, Henley Road, Medmenham,
PO Box 16, Marlow, Bucks, SL7 2HD
Telephone: Henley (0491) 571531

SUMMARY

(i) BACKGROUND

Utilisation on agricultural land is the major disposal outlet for sewage sludge in the UK, accounting for about 50% of the sludge produced annually. Metals in sludge applied to agricultural land remain in the cultivated layer of top soil. It is important that repeated applications of sludge do not build up metal concentrations in the soil to levels which could adversely affect crops or the animals, including man, which eat them. Field experiments were established at three sites on soils broadly representative of UK agriculture. Controlled applications of sludge were made and soil and crop concentrations of metals were monitored thereafter over a five year period. The results have defined the effect of sludge applications of known metal content on total and extractable concentrations of metals in soil. The results have defined also the extent of transfer of metals from sludge-treated soil into the leaves and edible parts of six crops of major importance to UK agriculture.

(ii) OBJECTIVES

Assist in the formulation of guidelines for the safe disposal of sewage sludge to agricultural land:

1. to determine the uptake of metals (particularly cadmium) by crops;

2. to determine the effect of soil type on the uptake of metals by crops;

3. to determine the yield response to the nutrients and potentially phytotoxic elements in the sludge;

4. to examine the mobility of trace-metal contaminants within the soil after sludge application.

(iii) PROGRAMME

Crops to be grown and harvested on an annual rotation over five successive seasons, with the following variables:

1. four types of digested sludge, each having different concentrations of cadmium;

2. six rates of application, including nil, to give the required range of metal levels in the soil;

3. six crops one being a long term ley of grass;

4. replication of the experimental treatments.

Yield from each plot to be measured, and analysis of the following to be made where appropriate, for nutrients, cation exchange capacity, organic matter, total metals (Ni, Cu, Zn, Cd, Pb) and available metals (Ni, Cu, Zn, Cd, Pb - by the methods of the Agricultural Development and Advisory Service of the Ministry of Agriculture, Fisheries and Food methods):

1. samples of the soil taken before the (once-only) application of the sludge;

2. samples of soil taken after sludging and rotavating, and thereafter annually;

3. samples of each crop taken at maturity, analyses being made on the edible parts of each plant and, where funds approved permit, on other selected parts.

(iv) EFFECTS OF SLUDGE ON SOIL CONCENTRATIONS OF METALS

1. The amount of cadmium applied in liquid sludge to the top 15 cm of three diverse soil types could be entirely accounted for by soil analyses in this profile, approximately five years after sludge application. The increase in soil metal (Mi) above background values was estimated according to soil density, cultivated depth, and metal loading rate as:

$$Mi = 10 \times Ml/(\rho \times s) \text{ mg/kg}$$

where Ml is the metal loading rate (kg/ha)
ρ the soil density (g/cm^3), air-dried
s the depth of cultivation (cm)

Where the weight of sludge applied exceeds ten percent of the weight of soil in the cultivated layer, then the effect of sludge on soil density should be taken into account.

Amounts of other metals, Ni, Cu, Zn and Pb applied in liquid sludge were also accounted for in the cultivated profile with the exception of Cu and Zn in the calcareous loam where some mobility may have occurred beyond 15 cm depth or there may have been some lateral movement of sludge-treated soil off the plots following cultivation.

2. The percentages of EDTA-extractable Ni, Cu, Zn and zinc equivalent (ZE) of total in unsludged soils were lower than for metals of sludge origin:

	unsludged soil			
	Ni	Cu	Zn	ZE
non-calcareous	13	39	6	13
calcareous	4	20	19	12

3. The percentages of EDTA-extractable Ni, Cu, Zn and ZE of total metal of sludge origin for the liquid and bed-dried sludges in non-calcareous and calcareous soil were as follows:

sludge	Liquid sludges				Bed-dried			
	Ni	Cu	Zn	ZE	Ni	Cu	Zn	ZE
non-calcareous	68	74	87	78	62	57	87	68
calcareous	22	37	46	33	12	27	26	21

Crop concentrations of the most plant-available metals (Cd, Zn and Ni) generally followed the pattern of percentage EDTA-extractability being highest on non-calcareous soils and liquid sludge treatments.

(v) EFFECTS OF SLUDGE ON CROP YIELDS

1. Crop yields were not significantly affected by sludge treatments in 60% of all cases studied.

2. Crop yields were significantly increased in 26% of cases of liquid sludge addition and this was attributed to the beneficial effects on soil

structure. Maximum dry solids additions to the soil from liquid sludge was 150 tonnes/hectare and 500 tonnes/hectare for bed-dried sludge.

3. Small (6-10% reductions) but statistically significant reductions in wheat grain yield were seen on the clay and calcareous loam soils treated with liquid sludge and the sandy loam and clay soils treated with bed-dried sludge. The results of comparisons between sludge types suggested this was not due to metals and the most likely explanation was lodging of the crop which occurred due to excessive nitrogen in the soil resulting from the single very heavy application of sludge made in the first year of the trial.

(vi) EFFECTS OF SLUDGE ON CROP COMPOSITION

1. Increases in metal concentrations in soil due to sludge added, produced significant increases in Cd, Ni, Cu and Zn concentrations in the edible portion of most of the six crops grown: wheat, potato, lettuce, red beet, cabbage and ryegrass.

2. Lead was relatively unavailable to crops from the soils. In the vast majority of cases there was no significant increase of Pb in crop tissue in relation to Pb added to the soil in sludge.

3. Increases in Cd concentrations in crops were most frequently directly proportional to the total Cd concentration in the soil. Thus they were estimated by a linear regression model fitted to Cd in crops plotted against 'total' Cd in soil. Increases in crop concentrations of Cd (mg/kg dm) per 1 mg/kg increase in Cd content of liquid sludge-treated soil (air dry) are summarised below (see also Tables 29 and 42):

Crop	Soil*	Estimated increase in crop tissue concentration per 1 mg/kg increase in soil content.
Wheat grain	sl	0.097
	c	0.105
	cl	0.046
Potato tuber	sl	0.042
	c	NS
	cl	0.028
Lettuce	sl	0.75
	c	1.2
	cl	0.34
Red beet root	sl	0.22
	c	0.24
	cl	0.07
Cabbage	sl	0.034
	c	0.051
	cl	0.044
Ryegrass	sl	0.084
	c	0.079
	cl	0.040

* sl = sandy loam, c = clay, cl = calcareous loam
NS = relationship not statistically significant

4. Increases in Zn concentrations in crops were most frequently proportional to the logarithm of the EDTA-extractable Zn concentration in the soil. Zn levels in crops were best estimated by fitting a logarithmic curve to data when plotted against EDTA-extractable Zn in soil

(vii) EFFECTS OF TIME

1. In general terms agreement between actual and predicted concentrations of metals in soil was best in the last two years of the trial. The explanation for this may be that the repeated cultivations gradually led to better mixing of sludge and soil and hence reduced the error associated with soil sampling.

2. There was little evidence that sludge metals were lost from the cultivated horizon except where sludge was originally incorporated below this depth. Apparent losses of less than ten percent of total Cu and Zn applied to the calcareous loam were indicated. Downward or lateral loss of sludge-treated soil from the plots as a result of cultivation could explain this effect.

3. The EDTA-extractable fraction of total Cu, Ni and Pb from the two non-calcareous soils increased with time.

4. Cadmium and Zn availability to crops over 5 years showed only three definite significant seasonal variations but less than ten percent of cases indicated definite trends over 5 years. Overall there was no trend in crop availability on any soil over 5 years. There was no trend of reducing plant availability of metals over the 5 year period.

(viii) EFFECTS OF SOIL CONDITIONS AND SLUDGE TYPE

1. The EDTA-extractable fraction of total Ni, Cu and Zn was less from the calcareous loam than the two non-calcareous soils. The relative differences were: Ni one fifth, Cu one half, and Zn four tenths.

2. The rate of increases in Cd concentrations in crops grown on non-calcareous soils was on average double the rate on calcareous soil. This was attributed to the difference in pH of 6.5 for non-calcareous soils in contrast to 8.0 for the calcareous soil.

3. Although the non-calcareous soils had the same pH, Cd was 1.6 times more available from the clay than the sandy loam to lettuce and cabbage. Even on unsludged treatments Cd was most available from the clay. The explanation for this may lie in the low content of iron and manganese sesquioxides of the clay soil.

4. Metals expressed as zinc equivalent were about 10% less extractable from soil treated with bed-dried sludge than from soil treated with liquid sludge (Table 40, Page 95).

5. The availability of metals to crops was in general terms lower in soil treated with bed-dried sludge compared with liquid sludge. This effect depended on the crop in question. Cadmium availability to crops from bed-dried sludge was found to be a half to an eighth of that from liquid sludges (Table 42, Page 97). This was probably because physically discrete bed-dried sludge particles remained in the soil during the trial reducing the likelihood of contact with crop roots. However, EDTA analysis indicated that metals may also be in less available chemical forms. Physical effects would be expected to decrease with time particularly on cultivated land.

(IX) RELATION TO CURRENT RECOMMENDATIONS FOR SLUDGE UTILISATION

1. Nickel, copper and zinc concentrations in soil produced by the upper rates of liquid and bed-dried sludge were close to the maximum permissible levels set out in the EC Directive for the use of sewage sludge in agriculture. In terms of additions of zinc equivalent, upper

rates of sludge application to soil exceeded the maximum permitted in UK guidelines by about 2x and 4x for liquid sludge and bed-dried sludge respectively (Table 34, Page 85). Nevertheless, no phytotoxic effects of metals were evident in terms of reduced yields, toxic tissue concentrations or visible symptoms. The only exception to this finding concerned concentrations of copper and zinc in lettuce grown on the clay soil which exceeded upper critical concentrations at high rates of sludge application.

2. In general terms there was no significant increase in crop concentrations of lead according to sludge treatment. Sludge applications did not influence the lead content of crops by the plant uptake route up to a soil concentration of Pb of at least 330 mg/kg, the highest recorded in the trial. This is the comparison to the EEC upper limit of 300 mg/kg.

3. In general terms there was a linear relationship between concentrations of cadmium in soil and concentrations of cadmium in the edible parts of the crops studied. An increase of 2.5 mg/kg in the soil concentration of cadmium produced the following increases in crop tissue content of cadmium (mg/kg dm):

Crop	Non-calcareous soils	Calcareous soils
Wheat grain	0.25	0.12
Potato tuber	0.11	0.07
Lettuce	2.4*	0.85
Red beet root	0.58	0.18
Cabbage	0.11*	0.11
Ryegrass	0.20	0.10

* see (viii) 3.

4. The percentage increase in crop cadmium content on calcareous soil was usually less than that seen on the two non-calcareous soils. Extractability of the zinc equivalent metals by EDTA from calcareous soils treated with sludge was about 40% of extractability from non-calcareous soils treated with sludge.

CONTENTS

Page

1. INTRODUCTION

During the late 1950s the practice of supplying liquid digested sludge to farmers and spreading it on their land was recognised as a particularly economic disposal option for inland sewage treatment works (Kershaw et al 1962). As the benefits to farmers were defined (Coker 1966) use of this outlet increased and was further encouraged by the Jaeger Report (MHLG 1970) which recommended that wherever possible encouragement should be given to the application to agricultural land of suitable sewage sludges. The question of metal accumulation in sludge-treated soils was well known and guidelines for copper, nickel, zinc and boron were set by MAFF (Chumbley 1971) and for 11 elements by the Report of the Working Party on the Disposal of Sewage Sludge to Land (1977). The latter report recommended that centrally co-ordinated research work, including field trials on the effects of metals, should be carried out to verify and refine the guidelines. It was with this in mind that fields trials at Cassington and Royston were started in 1977/78.

The trials consisted of three identical experiments based on a design agreed by expert consultants. Two experiments were set up at Cassington and managed by Thames Water and the third at Royston to be managed by the Water Research Centre. The sites chosen were on three contrasting soils broadly representative of UK agriculture. Each experiment included six blocks of plots so that six major crops could be grown continuously in rotation for the five year period of the trial. The plots were to receive graded applications of similar sludges with variable metal content so that the beneficial effects of sludge on crop yields, resulting from the effects of plant nutrients and organic matter, could be separated from the effects of metals. The experiment was concerned particularly to quantify the effect of cadmium in sludge-treated soil on the quality of food crops for human consumption. Current guidelines (DoE/NWC 1981) are such that cadmium is the metal which most often limits sludge applications to agricultural land. Metal limits for sludge utilisation need a scientific basis to ensure environmental protection without undue costs for sludge disposers or industry. The overall objective of the field trials was to contribute towards this scientific basis for metal limits. Utilisation in agriculture is currently the disposal outlet for about 50% of UK sludge.

This final report is a summary of approximately 50 000 results of soil and plant analyses carried out during the five year duration of the trial.

Progress reports listing all the results as they were obtained have been submitted throughout the period of the experiments and a list of these reports is included, including some interpretive reports; TW/WRc 'Part A' reports and Jolliffe 1981 and 1984.

2. METHODS

2.1 Experimental design

The trial was planned for 5 cropping seasons allowing a complete rotation of wheat, potatoes, cabbage, red beet and lettuce along with a ley of ryegrass. The design of the trial at each site was identical, consisting of 6 randomised blocks of 30 plots each 10 m x 1.8 m, making a total of 180 plots. There were 15 treatments within each block, comprising 5 rates of application (including a nil) of 3 sludges, all treatments being replicated in each block. Two of these original treatments were amended with a fourth sludge type during the trial (Watson 1979) at two rates of application. During the course of the trial the crops were grown on each block in rotation, except for ryegrass which remained on the same block throughout.

2.2 Site locations

One trial managed by the Water Research Centre, took place at Royston on a calcareous loam of high pH. The two remaining trials managed by Thames Water, were carried out at Cassington. The soils were a sandy loam and clay, both of low pH which was raised by liming to a level of 6.5 recommended for normal agricultural production (Chumbley 1981). The sandy loam had a low cation exchange capacity compared to the clay soil, as shown in Table 1. Details are also given of the total metal concentrations in the three soils before sludge was applied. Precise locations are shown in Appendix A.

Table 1. Details of the soils

Location	Royston	Cassington	
Texture	Calcareous loam	Sandy loam	Clay
Series	Swaffham Prior	Sutton	Carswell
Cation exchange capacity, CEC (meq/100 g)	20	13	29
pH value	8.0	6.5*	6.7*
Strong acid soluble metals (mg/kg)			
Cd	0.46	<0.25	0.65
Pb	45	21	26
Cu	18	20	23
Ni	9	32	40
Zn	56	99	125

* after liming (TW 1980a)

2.3 Sludges

Sludges S1 and S2 were similar in nature both being anaerobically digested and lagoon matured, although their metal contents differed. A third sludge (S3) was obtained by mixing sludge S1 and S2 in the ratio of 1:2 on a dry solids basis, for the calcareous loam and 1:3 for the sandy loam and clay at Cassington. Sludge was applied to the plots during the period early 1978 to March 1979. At Cassington earth banks were created around each plot before applying the sludge by metered pump. At Royston oil tempered hardboard was installed around each plot 300 mm above and below the soil level before the sludge was applied. Metal concentrations in the three sludges used at the two sites are given in Table 2.

Table 2. Concentrations of metals in the liquid (S1-3) and bed-dried (S4) sludges (mg/kg ds) and dry solids (ds%)

Element	Royston sludge type				Cassington sludge type			
	S1	S2	S3	S4	S1	S2	S3	S4
Cd	68	12	31	42	69	21	33	38
Pb	500	420	447	420	810	660	698	460
Cu	1160	400	650	760	1040	640	740	630
Ni	240	60	120	232	262	84	129	223
Zn	2050	1100	1420	1332	2310	1480	1690	1220
ds%	10	5-14	-	76	10.3	9.7	-	92

In 1979, after the first cropping season, it was decided to incorporate 2 further treatments into each block using a dried digested sludge (S4) which was essentially similar to S1 before being bed-dried (Watson 1979). The analysis of the bed-dried sludge is included in Table 2.

The treatments in terms of metal additions at the two sites have been adjusted to take account of small variations found amongst batches of sludge received on-site. Tables 3 and 4 detail the additions of dry solids and metals in kg/ha for Cassington and Royston respectively.

2.4
Crops

The six crops grown were selected to include major constituents of the human diet, metal-sensitive indicator crops and ryegrass. Approximately 70% of the plant dry matter intake of the average UK consumer is made up of bread, cereals and potatoes. Therefore winter wheat (*Triticum aestivum* cv Maris Huntsman) and potatoes (*Solanum tubersosum* cv Pentland Crown) were selected although due to poor crop establishment in the Autumn of 1978 and the continued sludge applications at Royston until March 1979 spring wheat (*cv Timmo*) was grown in 1979. Cabbage (*Brassica oleracea* cv Stonehead) was selected to represent the large number of green leafy vegetables consumed. The two indicator crops chosen were red beet (*Beta vulgaris* cv Crimson Globe) and lettuce (*Lactuca sativa* cv Mildura). Crops were grown in rotation on blocks B to F on all sites whilst block A was used for a long term ley of ryegrass (*Lolium perenne* cv Melle).

2.5
Plot establishment

Site selection and preparation was started at Cassington in 1977 when the two sites were selected on level ground at Worton Rectory Farm. The first on sandy loam was close to Yarnton Village on a gentle ridge of land to the east of the farm. The second, a heavy clay soil, was situated to the south of the farm buildings on level low ground. During the Autumn of 1977 180 plots were marked out at each site followed by comprehensive soil sampling. The arrangement of plots within blocks

Table 3. Metals added to soil by the sludge treatments on sandy loam and clay

Sludge type	Treatment	Dry solids addition (t/ha)	Metal additions (kg/ha) Cd	Pb	Cu	Ni	Zn
S1	R1	19	1.3	16	20	5.3	44
	R2	38	2.6	31	41	10.5	88
	R3	76	5.3	63	81	21	177
	R4	153	10.5	125	162	42	353
S2	R1	18	0.4	12	12	1.6	27
	R2	36	0.8	24	24	3.1	53
	R3	72	1.5	48	47	6.2	106
	R4	144	3.0	95	94	12.4	212
S3	R1	18	0.6	13	14	2.5	31
	R2	37	1.2	26	28	5.0	62
	R3	74	2.5	52	56	9.9	124
	R4	147	4.9	103	111	19.8	247
S4	R4	233	8.8	107	147	51.8	285
	R5	466	17.6	214	294	104	570

Table 4. Metals added to soil by the sludge treatments on calcareous loam

Sludge type	Treatment	Dry solids addition (t/ha)	Metal additions (kg/ha) Cd	Pb	Cu	Ni	Zn
S1	R1	19	1.3	9.5	22	4.5	39
	R2	38	2.6	19	44	9	78
	R3	76	5.2	38	88	18	156
	R4	152	10.4	76	176	36	312
S2	R1	19	0.24	8	7.5	1.2	21
	R2	38	0.48	16	15	2.4	42
	R3	76	0.95	32	30	4.7	83
	R4	152	1.90	65	60	9.3	166
S3	R1	18	0.6	8.5	12	2.3	37
	R2	35	1.2	17	24	4.5	53
	R3	71	2.3	33	48	9	105
	R4	141	4.6	66	96	18	210
S4	R4	250	10.5	105	190	58	333
	R5	500	21	209	380	116	666

varied between sites to accommodate the experiment within topographic limitations. After the plots were marked out earth banks were created approximately 300 mm high around each plot to retain the applied sludge. Sludge applications took place intermittently during 1978 according to weather conditions, but were completed by August. The plots were cultivated as soon as drying took place and weeds were controlled where necessary with paraquat herbicide.

At Royston work commenced in the Autumn of 1978, with site selection and detailed background soil analyses. A trencher was used to cut a 100 mm wide slit in the soil 300 mm deep along all plot boundaries. Oil tempered hardboard was inserted in the soil with 300 mm standing above the soil surface. During the Winter of 1978/79 digested sludge was pumped into these plots; using a Molex pump of known output. As at Cassington this was allowed to run for a period of time according to the sludge additions required for individual treatments. Sludge applications were completed by March 1979.

At both sites the sludge was allowed to dry partially before hand digging to incorporate the sludge within the soil. When the sludge was completely dry the plots were cultivated using either a tractor-mounted or hand-operated rotary cultivator.

Once seed beds had been prepared the cropping sequence was started. At Cassington there was a period of equilibration of 1 year between sludge applications and cropping whilst at Royston only a few weeks elapsed. The sludge was therefore relatively freshly applied compared to the DoE guidelines 1981 which refer to a 30 year build up of metals in the soil (DoE/NWC 1981).

2.6

Plot maintenance and farming practices

The production of all crops followed good farming practice as far as possible. There was an unsuccessful sowing of the lettuce and ryegrass during 1978 but in practical terms all three sites started cropping in the Spring of 1979. There were several differences in individual operations and timing to suit soil conditions at each site. At Royston the calcareous loam over chalk faced south and soon dried in the spring allowing early cultivations with tractor-mounted equipment. The tractor wheels were set wide apart so that the tractor wheel centres coincided with the plot boundaries. The cultivation equipment was narrower than the wheels and therefore a narrow strip of soil was uncultivated which formed the plot boundary. The depth of cultivation was carefully controlled by the tractor three point linkage. Usually one pass of a springtine cultivator was sufficient at Royston to produce a suitable seed bed for winter wheat. A second pass of the springtine cultivator was sometimes needed for the small seed crops lettuce, cabbage and red beet. Consolidation was usually necessary using a spiral or Cambridge roller depending upon soil conditions. At Cassington a hand operated garden cultivator was usually used and two or more passes were often necessary to prepare suitable seed beds. The plots were then raked by hand to effect consolidation and improve uniformity of the surface. Small seed crops at all sites were sown using a single or five row Stanhay precision drill. The winter wheat at Royston was broadcast whilst at Cassington it was drilled. The potato crops were planted by two row machine at Royston and by hand at Cassington. The ryegrass was broadcast at all sites.

Inorganic fertiliser was used on all crops in accordance with MAFF recommendations (1980, 1981a), and are summarised in Table 5. A variety of straight nitrogen, phosphorus or potassium fertilisers along with compound fertilisers were used during the experiment.

Table 5. Mean fertiliser applications each season (kg/ha)

Crop	Nitrogen (N)	Phosphate (P_2O_5)	Potash (K_2O)
Wheat	120+	60	60
Potato	210	260	250
Lettuce	120	170	150
Red beet	200	100	180
Cabbage	210	125	225
Ryegrass*	110	50	50

* per cut
+ includes top dressing

At Royston and Cassington a limited number of chemical sprays were used. The plots were usually treated with paraquat before sowing to kill any weeds present and again after harvesting, although glyphosate herbicide was used when perennial weeds were present. Some herbicides were used on the growing crop when necessary particularly for potatoes. On the sites at Cassington paraquat and glyphosate herbicides were used with similar regularity as at Royston. However more use of herbicides and pesticides was made at Cassington on all crops. Despite the use of herbicides on the experiment hand weeding proved to be a major task during the early summer months at both sites.

Details of farming practices are given in Appendix B.

2.7 Sampling procedures

The sampling procedures followed were similar at all three sites. Soil core samples were taken 0-15 cm at the beginning of each year. The sampling frequency was 15 cores per plot at Royston but at Cassington 25 per plot were taken in 1979 and 20 in subsequent years. Depth samples were taken on selected blocks in certain years to compare sludge incorporation depth and leaching of metals down the profile at each site. All soil samples were dried at less than 30 ^{o}C before grinding to pass a 2 mm sieve prior to analysis.

Crop samples were taken before harvesting at a growth stage suitable for marketing the crop. The ryegrass plots were cut and the grass subsampled after weighing. Samples of winter wheat heads were taken from the standing crop for analysis before harvesting and measuring yields of grain and straw. A sub sample of straw was then taken for analysis. The extent of lodging (flattening) of wheat was also assessed by scoring prior to harvesting.

The sampling of the row crops lettuce, red beet and cabbage followed the same procedure. Fifteen plants were selected at random during harvesting for analysis. The fresh yield of the 15 plants and the rest of the middle three rows was determined. The lettuce and cabbage were washed and quartered before drying at 70 ^{o}C and grinding to pass a 1 mm sieve. The red beet were initially quartered but subsequently the roots were peeled before quartering and drying. Two rows of potatoes were grown on each plot and harvested mechanically, 15 tubers were selected at random for washing, peeling and drying before grinding. For potatoes and red beet both the peels and inner tubers were analysed.

2.8 Chemical analysis

Flame atomic absorption spectrometry was the method used to detemine metal concentrations in plant and soil samples. Different methods of preparation were used for the Cassington and Royston sites. Samples from Cassington were digested in nitric/perchloric acids, and were made up to volume using hydrochloric acid. For soils the ratio of final volume to weight was 25 (eg 1 g to 25 ml) and for plant materials 10 (eg 2.5 g to 25 ml). MAFF methods were followed (MAFF 1978). Royston samples were digested in nitric acid only, the procedure differing between plant materials and soils only in the weight of material taken, that is 0.5 g for soils and 1.25 g for plant materials. The methods for the Royston samples are given in Appendix F. Comparability of results between laboratories was checked by exchange of samples throughout the period of the trial and by the use of standard reference materials. During the trial a new method was recommended for determination of Cd in plant material (MAFF 1981b). In view of the low concentrations encountered for Cd in crop samples grown on unsludged soil, samples of wheat and potato were reanalysed by the new method (Appendix F). The reanalysis confirmed the original results for Cd in wheat grain. For potato, however, differences were significant and attributed to difficulty in establishing the true value of the blank. They were not sufficient to influence the overall crop-soil relationship determined. Extractable metals were determined by MAFF (1976) methods for all soils.

2.9 Statistical methods

An analysis of variance was carried out on all soil and crop data on a block by block basis. Residual errors, differences between observed and fitted values, were examined to ensure there was no pattern within each block, that is, fertility trends were not present. The residuals were also examined in relation to the sludge rate to confirm values were approximately constant to justify assumptions made in significance tests and regression analyses. Outlier values were identified on a statistical basis, once again by observing residuals and comparing them to their standard error.

In analyses of variance, significance of observed effects within blocks was determined from the model:

$$Y_{ij(kl^{n})} = (GM + E_{ij} + (S_k + R_{lk} + T_{nk} + SRT_{kln}) \quad (1)$$

where

$Y_{ij(kln)}$	observed value of Y in row i and column j
GM	mean term of all values
E_{ij}	residual effect of row i in column j
S_k	main effect of sludge at level k
R_{lk}	main effect of rate at sludge k and level l
T_{nk}	main effect of type at sludge k and level n
SRT_{kln}	effect (interaction) of the combination ln of rate and type at sludge level k

More complicated comparisons involving several blocks, for example, when testing effects between years, were made by extending this basic model. Tukey's least significant difference (LSD) was calculated to test differences between treatment means at the 5% level of significance.

Correlations between parameters were measured by calculation of the Pearson's product moment correlation coefficient (r). The percentage variance accounted for by a regression (V%) was calculated as:

$$V\% = 100\ (\text{total MS} - \text{residual MS})/\text{total MS} \qquad (2)$$

where MS is the mean square of the appropriate sums of squares of the regression.

V% is a better descriptive statistic than r. Linear regression models were derived by the method of least-squares.

3. RESULTS

3.1 List of experimental results obtained

Many samples were analysed for extra determinands and additional samples were taken for analysis beyond those required by the contract when funds permitted. The required analyses were total and ethylenediaminetetra-acetic acid (EDTA) diammonium salt-extractable Cd, Ni, Cu, Zn and Pb in soil from each plot each year. Total Cd, Ni, Cu, Zn, Pb and yield in edible crop samples each year. Also required were necessary sludge and soil analyses before sludge application.

Further analyses that were carried out in addition to those specified in the contract are listed in Appendix E.

3.2 Metal concentrations in soils

Soil samples collected after each of the five cropping seasons were determined for strong acid soluble (total) metals and EDTA-extractable metals. Metals were cadmium, nickel, copper, zinc and lead. All plots were sampled, except in 1979 on the sandy loam and clay, when only blocks D, E and F were examined.

3.2.1 Effects of sludge application on soil concentrations of metals

Levels of metals added to the soil in the sludges were given in Tables 3 and 4. To discover how these loading rates had affected the soil metal concentrations, scatter plots were examined for each metal, noting sludge type, on a yearly basis. Effects of the three liquid sludge types in each year were similar as expected. The bed-dried sludge, however, was a different case as it was applied later than the liquid sludges (Section 2.3) and it was physically quite different with a very high percent dry solids (Table 2).

The relationship between metal loading rates and increase in metal concentration in soil as expected was linear with highly significant correlation. Regression analyses were used to estimate statistics describing the relation for the liquid sludges. Correlation coefficients are detailed in Appendix C. Figure 1 illustrates an example for Cd in 1980 by which time bed-dried sludge treatments had been added to the design. Points on the figure are for mean values for each sludge treatment. The regression of soil concentrations (individual replicate data, excluding bed-dried sludge) upon sludge metal loading rates is illustrated by a fitted line. The high correlation between the two parameters is indicated by the correlation coefficient r, and also by the percent variance accounted for by the regression V%. The soil concentration at zero loading rate is given (a) the best estimate of background soil metal in that

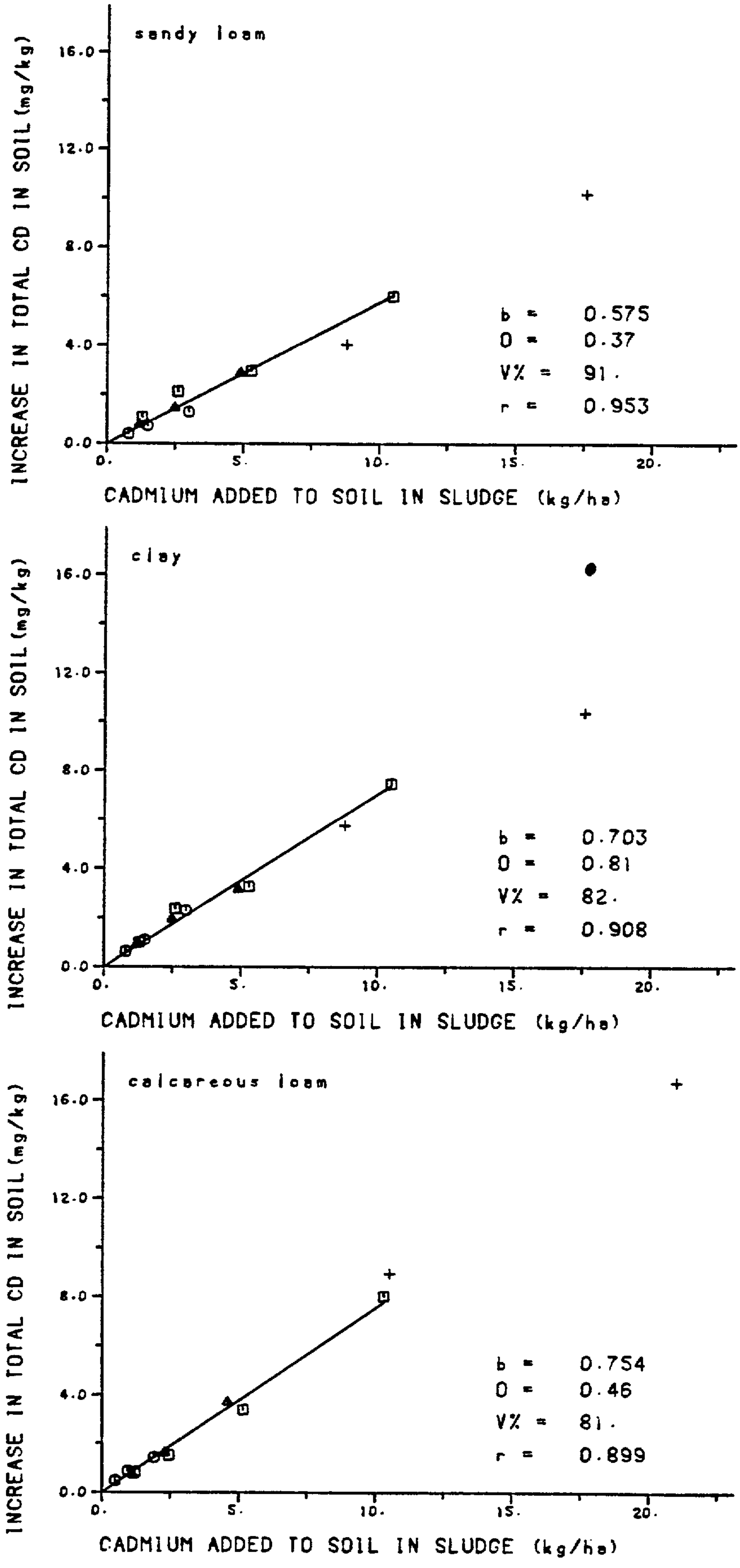


Fig 1. Cadmium increase in soil vs. cadmium added to soil. 1980

year, and this was taken as the origin above which inreases are measured (y-axis). The slope estimate b, for each soil gives the ratio of increase in soil cadmium (mg/kg) to cadmium added in the liquid sludges to soil (kg/ha). In 1980, this was three years after these liquid sludges had been applied on the sandy loam and clay and two years after for the calcareous loam. Theoretically the rise in soil metal (Mi) could be estimated as:

$$Mi = 10 \times Ml/(\rho \times s) \text{ mg/kg} \qquad (3)$$

where Ml is the metal loading rate (kg/ha)
ρ the soil bulk density (g/cm^3),
s the cultivated and sampling depth (cm)

Soil densities were measured by the 'excavation' method in 1980-83 (TW 1981-3a, TW 1984, WRc 1980b). From these data mean densities of treatments from which b was estimated, for the sandy loam, clay and calcareous loam were calculated as 1.38, 1.02 and 0.99 respectively. Thus, expected values of the ratio b after sludge application assuming no subsequent loss of sludge metal from the cultivated depth s would be 0.48, 0.65 and 0.67. These are somewhat lower than those found for cadmium in 1980 (Figure 1). The same relationships are given for all metals in 1980, for each site in turn, in Figures 2-4. Here there was much better agreement for Cu, Zn and Pb on the sandy loam and clay, also for Cu and Zn on the calcareous loam. Greatest accuracy is expected from data for Cu and Zn because their loading rates are highest and they are determined with better accuracy than the other metals. One factor producing high values for b could be sludge not yet incorporated to the cultivation depth.

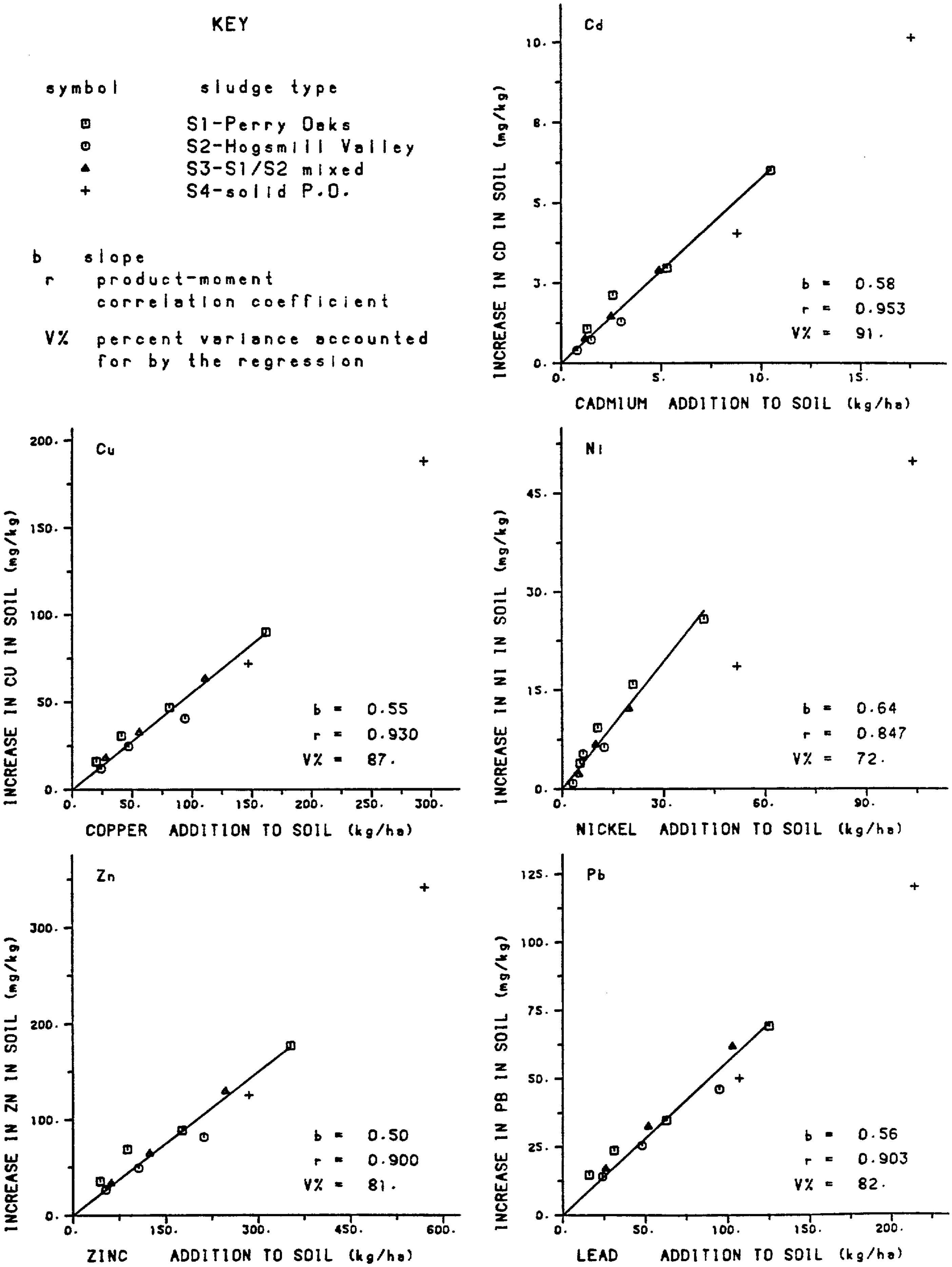


Fig 2. Metal increase in soil vs. metal added to soil. sandy loam 1980

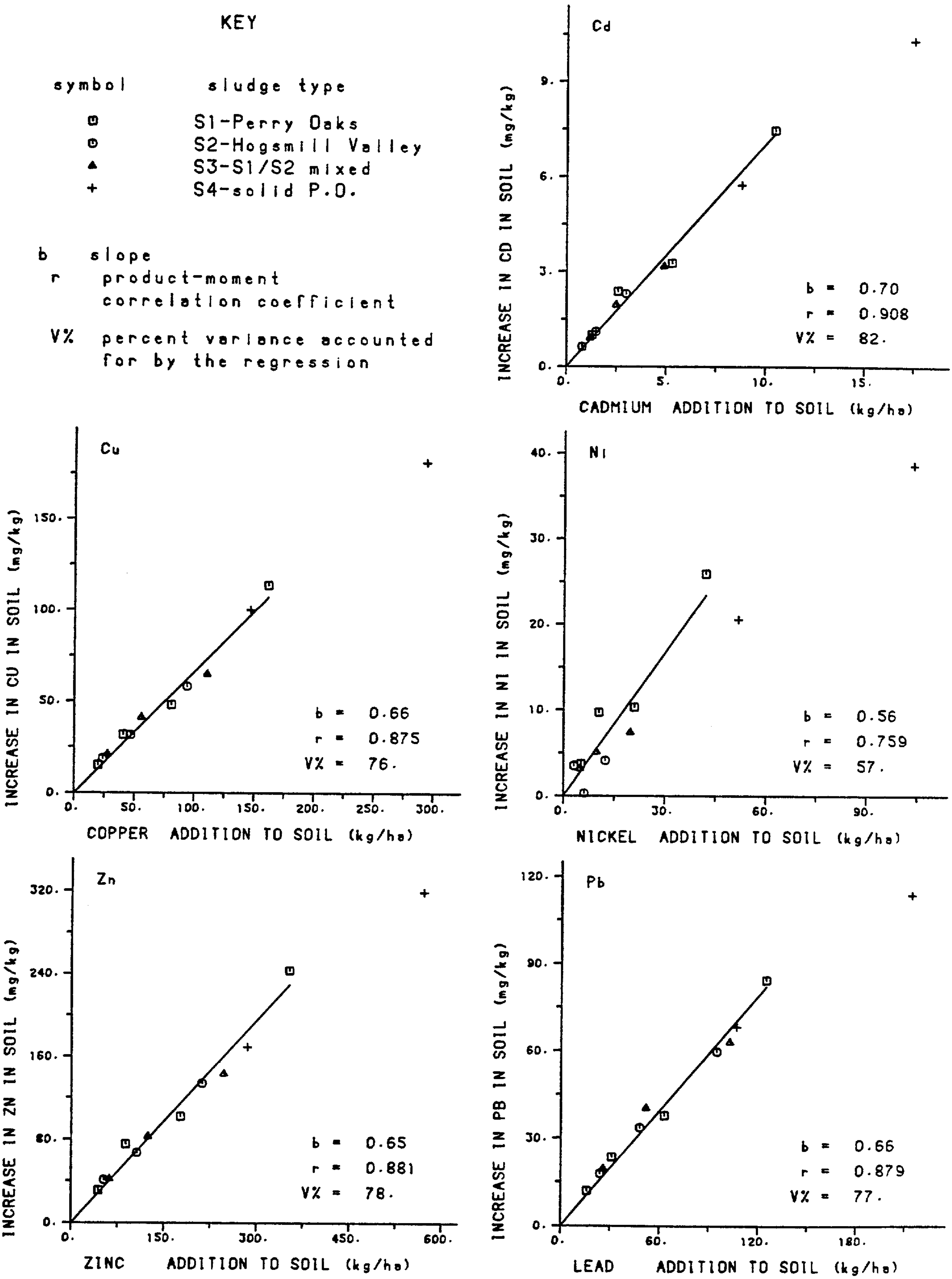


Fig 3. Metal increase in soil vs. metal added to soil, clay 1980

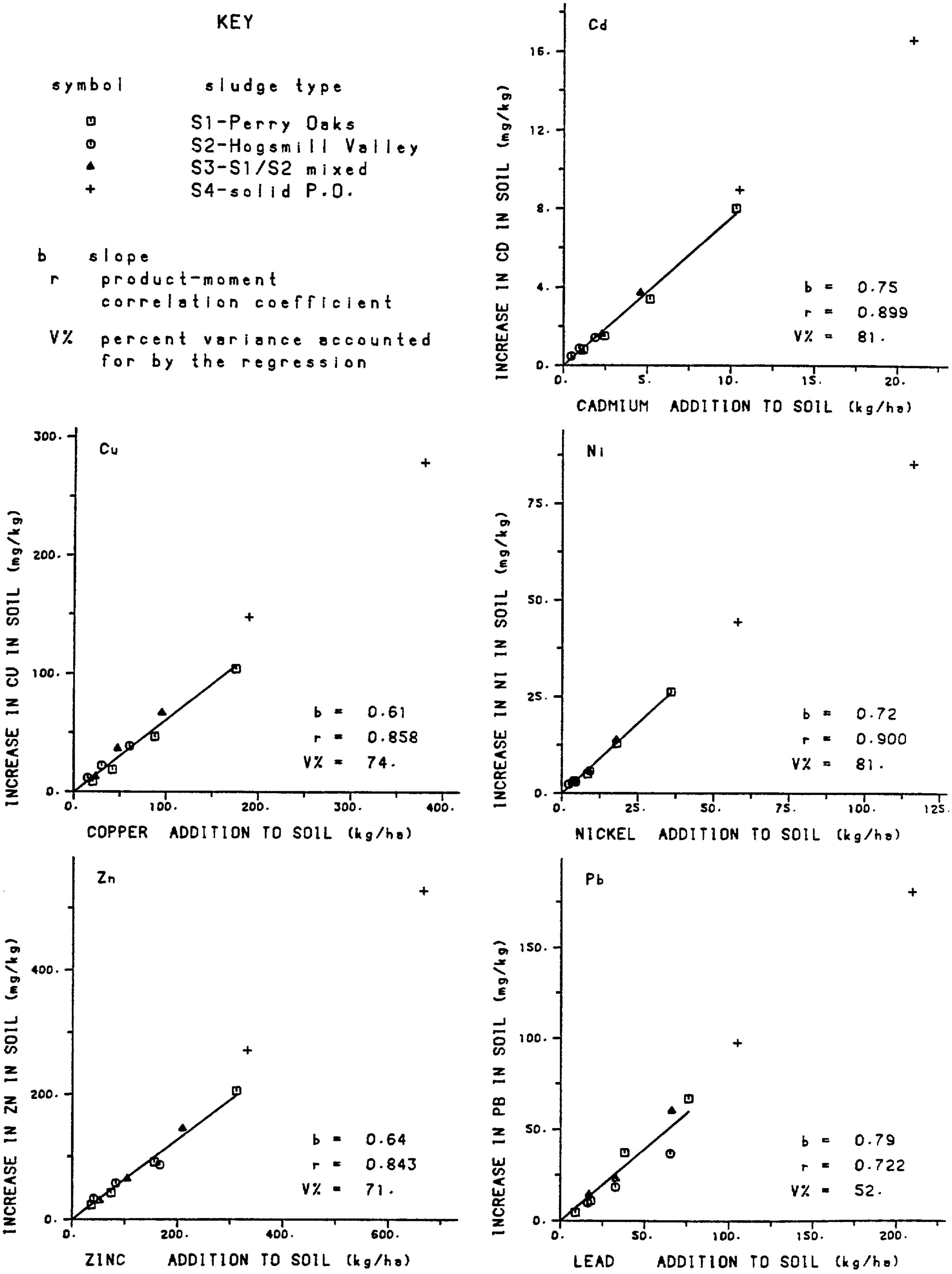


Fig 4. Metal increase in soil vs. metal added to soil. calcareous loam 1980.

Table 6. Relationship between Cd concentration in soil and Cd added in sludge to soil, liquid sludge treatments

Soil		1979	1980	1981	1982	1983
sandy loam	slope (b)	0.61	0.58	0.59	0.46	0.52
	SE	0.015	0.015	0.010	0.011	0.013
	constant (a)	0.21	0.37	0.30	0.30	0.29
	SE	0.052	0.057	0.039	0.041	0.048
	n	90	156	156	156	156
clay	b	0.82	0.70	0.77	0.54	0.67
	SE	0.030	0.026	0.027	0.025	0.020
	a	0.80	0.81	0.79	0.77	0.83
	SE	0.106	0.100	0.104	0.095	0.077
	n	90	156	156	156	156
calcareous loam	b	0.85	0.75	0.62	0.58	0.52
	SE	0.028	0.030	0.016	0.019	0.023
	a	0.65	0.46	0.72	0.91	1.08
	SE	0.094	0.108	0.059	0.070	0.083
	n	180	156	155	156	155

units : Cd concentrations in soil mg/kg (Y), Cd added to soil kg/ha (X)
slope : regression coefficient for Y upon X
constant : Y axis intercept
SE : standard error of slope or constant
n : number of points in regression

3.2.1.1 Effects between years

To illustrate the relation between Cd loadings to soil and increase in Cd in soil during five years of the trial 1979-83 for the liquid sludge treatments, fitted lines are given for each year in Figure 5. The statistics for these fitted lines are given in Table 6. Standard errors (SE) for each estimate are included. The 95% confidence interval for each estimate is approximately twice the SE. Values of b for the sandy loam and the clay for Cd appeared to drop overall towards the expected values from equation (3). Background soil estimates (a) were steady indicating a true reduction in soil Cd concentration within the liquid sludged plots. Thus it would appear sludge metal was gradually mixing towards the cultivated

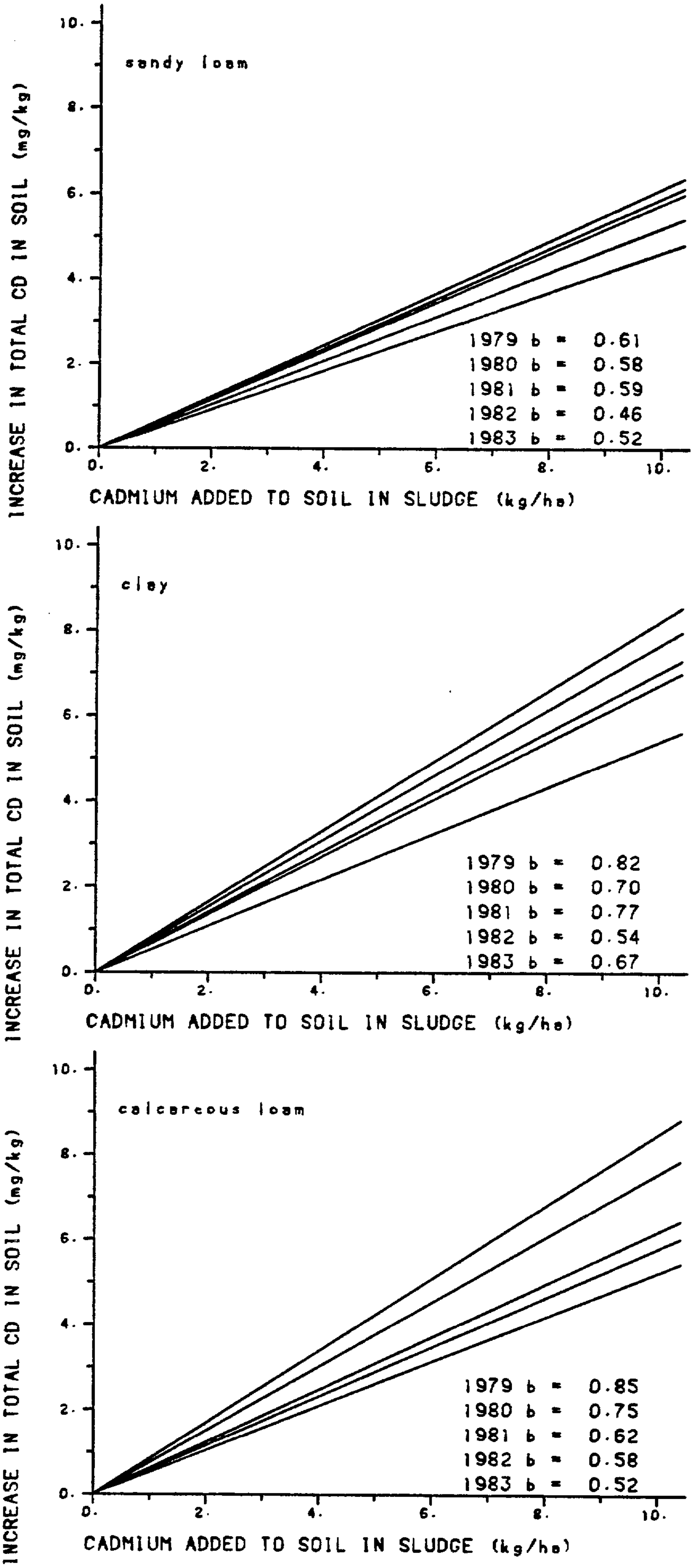


Fig 5. Cadmium increase in soil vs. Cadmium added to soil, 1979-83
(liquid sludge treatments)

15 cm depth with time. Larger reductions in b are seen for the calcareous soil, falling below that expected by 1981, however, this apparent mobility of Cd was not observed in the mass balance approach (Section 3.2.1.2). Regression statistics for Ni, Cu, Zn and Pb are given in Appendix C.

In order to establish the significance of changes in soil metal concentrations between years analyses of variance were carried out on concentrations in background and liquid sludged treatments, examining overall means for each year. A separate comparison for 1979-80 was made for the sandy loam and clay, including just three blocks D, E and F, see Section 3.2. The mean concentrations for all metals are given in Table 7. Where the effect between years was significant ($P \le 0.05$), the LSD is included. The significant effects confirm the differences indicated in the regression analyses each year.

Table 8 summarises mean Cd concentrations in background soil, liquid sludge treatments and bed-dried sludge. Soil densities for liquid and bed-dried sludge treatments were calculated once more (Section 3.2.1) and used to estimate theoretical mean concentrations (e) assuming no metal mobility (Equation 4). Densities for the bed-dried sludge treatments were significantly lower than those for liquid sludge, 1.22, 0.95 and 0.83 g/cm^3 for the sandy loam, clay and calcareous loam respectively. The concentrations for bed-dried sludge on all soils show an overall drop in a similar pattern to that seen for liquid sludge. The pattern on the sandy loam and clay was; approximately constant levels 1979-81 followed by a sharp drop in 1982 then rising in 1983 to a level which was still lower than that in 1981. For the calcareous loam Cd levels fell between the

Table 7. Mean total metal concentrations in soil, effects between years (background soil and liquid sludge treatments) mg/kg

Metal	Soil	1979	1980	1981	1982	1983	LSD 79/80,80/83
Cd	sl	1.85	1.94, 1.89	1.88	1.54	1.69	NA,0.14
	c	3.00	3.06, 2.70	2.86	2.24	2.66	NA,0.27
	cl	2.74	2.30	2.22	2.31	2.32	0.30
Ni	sl	42.7	44.5 ,42.3	40.6	37.5	40.4	1.41,1.17
	c	49.5	50.2 ,50.0	49.8	45.0	49.5	NA,1.41
	cl	19.7	17.5	18.5	17.1	17.3	1.30
Cu	sl	51.3	53.9 ,52.3	51.3	46.9	49.3	2.65,2.53
	c	65.8	67.6 ,61.9	65.5	52.7	62.0	NA,4.78
	cl	54.8	46.3	44.6	44.2	42.0	5.07
Zn	sl	198	186 ,182	184	155	168	7.9 ,6.5
	c	238	226 ,216	230	196	217	NA,9.9
	cl	145	128	121	119	121	11.1
Pb	sl	58.5	60.6 ,58.3	59.1	52.7	55.9	NA,3.1
	c	72.4	69.9 ,65.2	70.2	55.7	65.5	NA,4.38
	cl	78.5	76.7	74.7	69.8	68.9	6.04

soil type : sl - sandy loam, c - clay, cl - calcareous loam
treatments excluded : S1R1 and bed-dried sludge
note : the means for 1979 and the first mean for 1980 on soils sl and c are from soil analyses of Blocks D, E and F only (72 plots/year). The comparison for these by analysis of variance was separate to the comparison for 1980-83 where all 6 Blocks are included (144 plots/year)
LSD : the least significant difference between mean concentrations, given for each comparison at the 5% level of significance if applicable, otherwise; NA.

Table 8. Mean concentrations of Cd in soil, background soil, liquid and bed-dried sludge treatments and theoretical means (mg/kg)

Soil	Sludge		1979	1980	1981	1982	1983	e*
sandy loam	none	mean	0.31	0.26	0.29	0.27	0.38	
		n	18	36	36	36	36	
	liquid	mean	2.36	2.43	2.41	1.96	2.13	2.01
		n	54	108	108	108	108	
	bed-dried	mean	NA	7.47	7.31	4.73	5.97	7.47
		n	NA	24	24	24	24	
clay	none	mean	0.74	0.63	0.72	0.67	0.76	
		n	18	36	36	36	36	
	liquid	mean	3.75	3.39	3.57	2.76	3.29	3.04
		n	54	108	108	108	108	
	bed-dried	mean	NA	8.87	8.44	6.17	6.75	9.90
		n	NA	24	24	24	24	
calcareous loam	none	mean	0.62	0.59	0.64	0.76	0.99	
		n	36	36	36	36	36	
	liquid	mean	3.45	2.87	2.75	2.83	2.76	2.67
		n	108	108	108	108	108	
	bed-dried	mean	NA	13.30	10.76	10.00	10.18	13.23
		n	NA	24	24	24	24	

n number of plots/analyses
NA not applicable
e* theoretical concentration based on sludge additions (Tables 4 and 5), background soil metals before sludging (Table 1), incorporation to 15 cm depth and soil density (Equation (4)).

first two years of sampling and then remained approximately constant. Significance testing between years was not performed on the bed-dried sludges.

3.2.1.2
Mass balance for cadmium in 1983

Levels of Cd remaining in the cultivated depth in 1983 can be compared for both liquid and bed-dried sludge treatments with the theoretical values given in Table 8. The theoretical e values were calculated as:

$$e = Mi + STo \text{ (mg/kg)} \qquad (4)$$

where Mi is the theoretical rise in metal concentration due to sludge metal added, see Equation (3).

STo mean concentration of total metal in unsludged soil

The effect of sludge on soil density was taken into account. This was particularly important for the bed-dried sludge treatments where the weight of sludge applied was 10-23% of the soil in the cultivated layer.

For the liquid sludges on all soil types all Cd added by the sludge additions can be accounted for in the soil profile sampled in 1983, 4.5-5.5 years after sludge addition. This reinforces evidence from the relations between total Cd and Cd additions to soil seen in Section 3.2.1.1.

Mean Cd concentrations found in bed-dried treatments in 1983 fell well below the theoretical values on all soils. More than would be expected through sampling and analytical errors. This is the only evidence of loss of Cd from the plough depth and was not tested statistically. Tables of yearly means for Ni, Cu, Zn and Pb on the same basis as in Table 8 are given in Appendix D.

3.2.2 Relation between total and EDTA-extractable concentrations

There was a highly significant correlation, for each metal, between total concentration in the soil and EDTA-extractable concentrations. Total metal concentrations were measured by strong acid, see Section 2.8. Scatter plots of these two parameters for each metal/year combination together with correlation statistics, revealed linear relationships with good agrement between liquid sludge types. The bed-dried sludge treatments were distinct from the liquid sludge types and are considered separately in Section 3.2.2.1. Regression analyses were carried for the liquid sludge treatments and measures of correlation (r) are detailed in Appendix C.

Figure 6 illustrates an example for Cu determinations in 1980 on all three soils. The points are from treatment mean values, however, the regression analyses were based on the individual replicate data. Furthermore because the X-variate, total metal, had measured errors comparable to EDTA determinations a simple Y upon X regression was not appropriate (Ellis 1985). Instead for all relationships between EDTA and total metals in soil two models were initially fitted treating each parameter as independant in turn; Y upon X then X upon Y. The geometric average of the two then gives the estimates of the regression coefficients given in the figures for this relation.

In Figure 6 the bed-dried sludge is clearly distinct from the liquid types, the proportion of

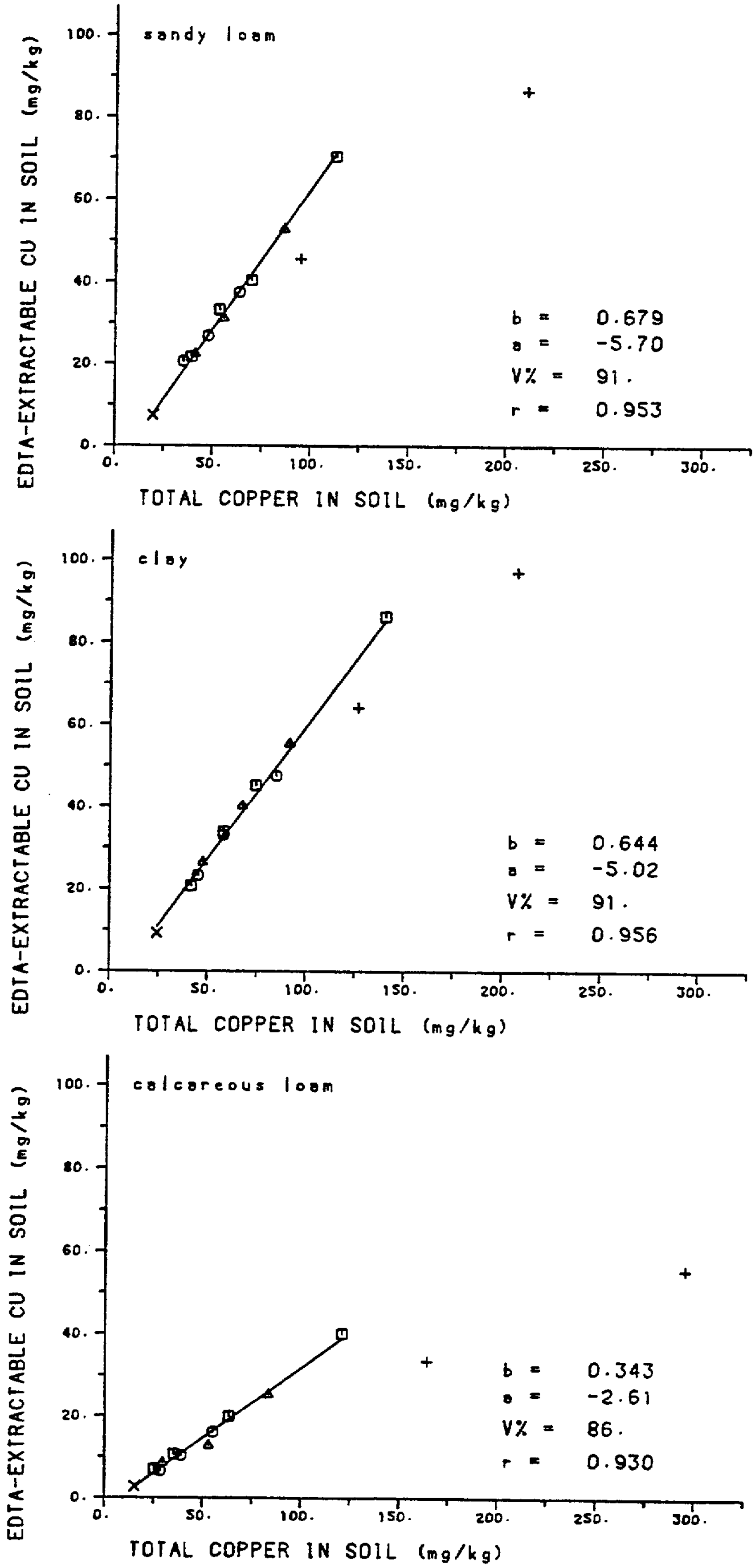


Fig 6. EDTA-extractable Cu in soil vs. total Cu in soil. 1980

Cu extracted by EDTA of the total was less. Hence the fitted line was estimated excluding the bed-dried sludge treatments. This was also true for other metals which are shown for each soil in turn in 1980 in Figures 7-9. There was little difference between the sandy loam and clay soils for any metal, however, amounts of Cu and other metals extracted by EDTA from the calcareous loam were much lower. In 1980 for the calcareous loam the proportion of Cu, Zn and Cd extracted by EDTA of the total was approximately half that found for the non-calcareous soils. Amounts of Ni and Pb were still lower; approximately a third.

3.2.2.1 Effects between years

The pattern seen in 1980 between soils is reflected in other years and models illustrating the relation for Cu for each soil are given in Figure 10 with further details in Table 9. Regression statistics for Ni, Zn, Cd and Pb in every year are detailed in Appendix C.

Table 9. Relationship between EDTA-extractable Cu and total Cu in soil, background soil and liquid sludge treatments

Soil		1979	1980	1981	1982	1983
sandy loam	slope (b)	0.63	0.68	0.785	0.81	0.89
	SE	0.022	0.017	0.0085	0.022	0.020
	constant (a)	-6	-5.7	-9.3	-9	-12
	SE	1.2	0.97	0.48	1.1	1.1
	n	90	156	156	156	156
clay	b	0.58	0.64	0.71	0.81	0.85
	SE	0.014	0.015	0.012	0.018	0.010
	a	-3	-5.0	-6.8	-8	-11.6
	SE	1.0	1.1	0.86	1.1	0.69
	n	90	156	156	156	156
calcareous loam	b	0.188	0.343	0.410	0.52	0.46
	SE	0.0054		0.0093	0.015	0.011
	a	0.9	-2.6	-4.0	-4.8	-4.0
	SE	0.34		0.47	0.72	0.52
	n	178	156	155	156	155

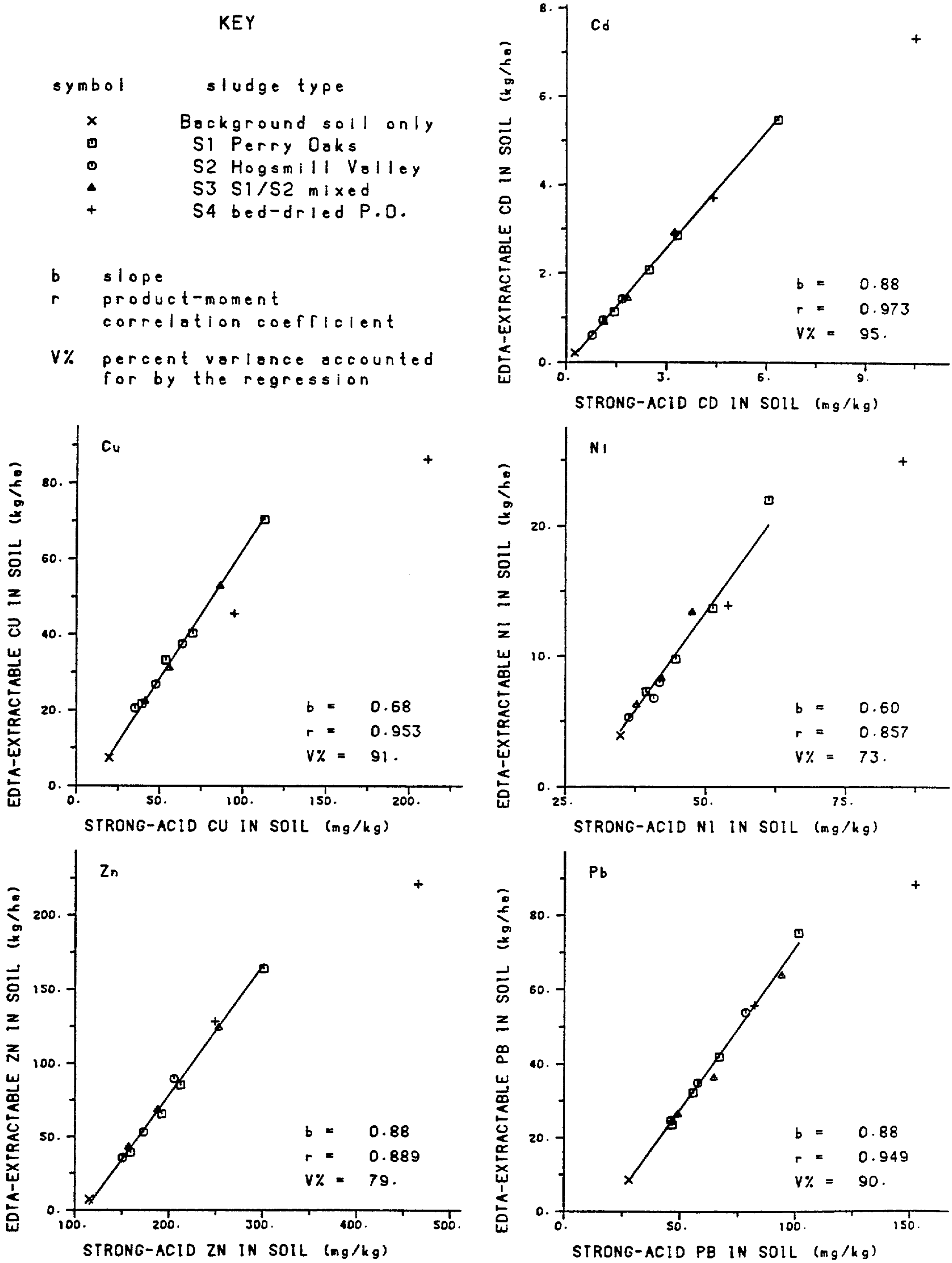


Fig 7. EDTA-extractable metal in soil vs. total metal in soil. sandy loam 1980

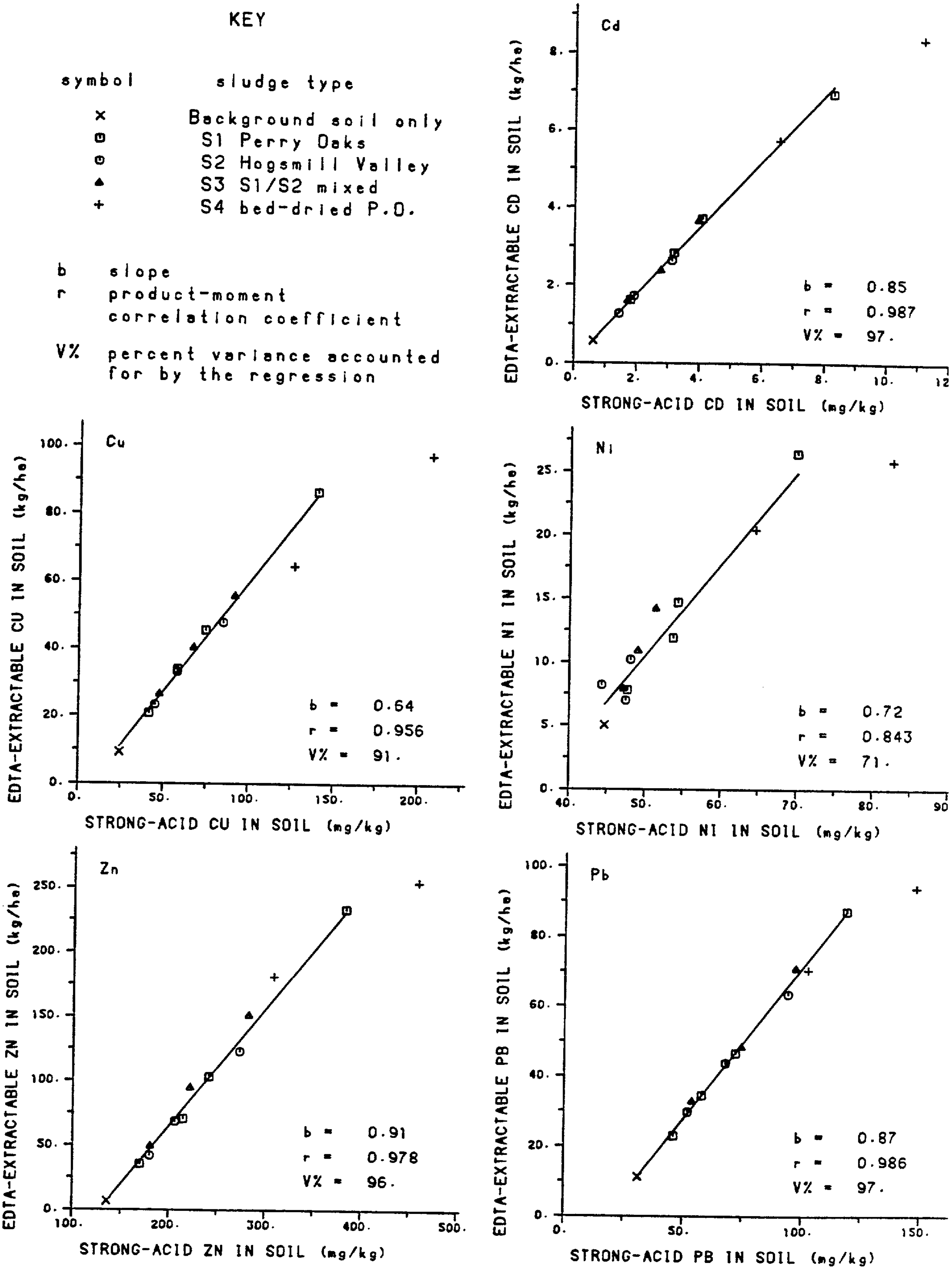


Fig 8. EDTA-extractable metal in soil vs. total metal in soil. clay 1980

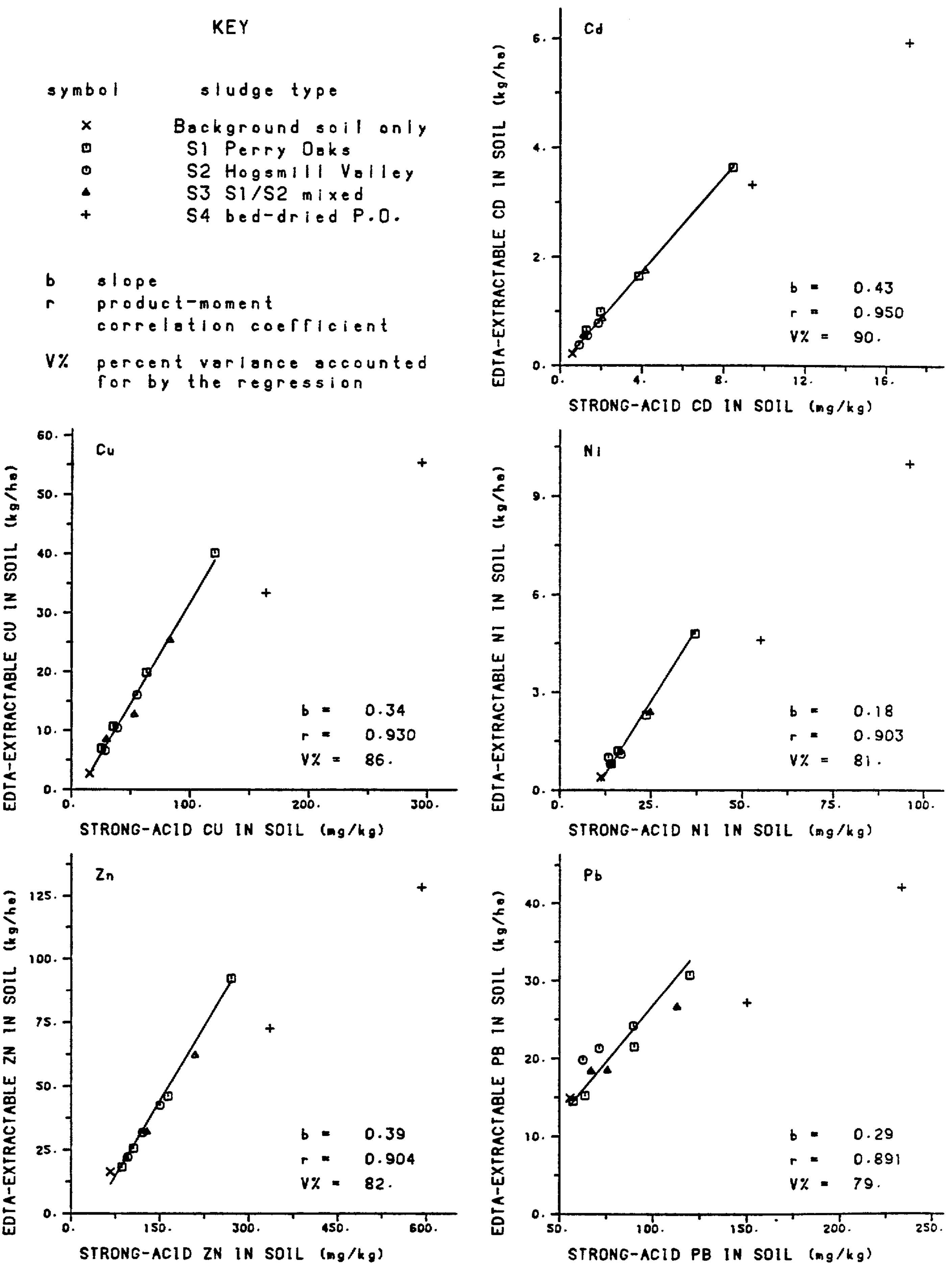


Fig 9. EDTA-extractable metal in soil vs. total metal in soil. calcareous loam 1980

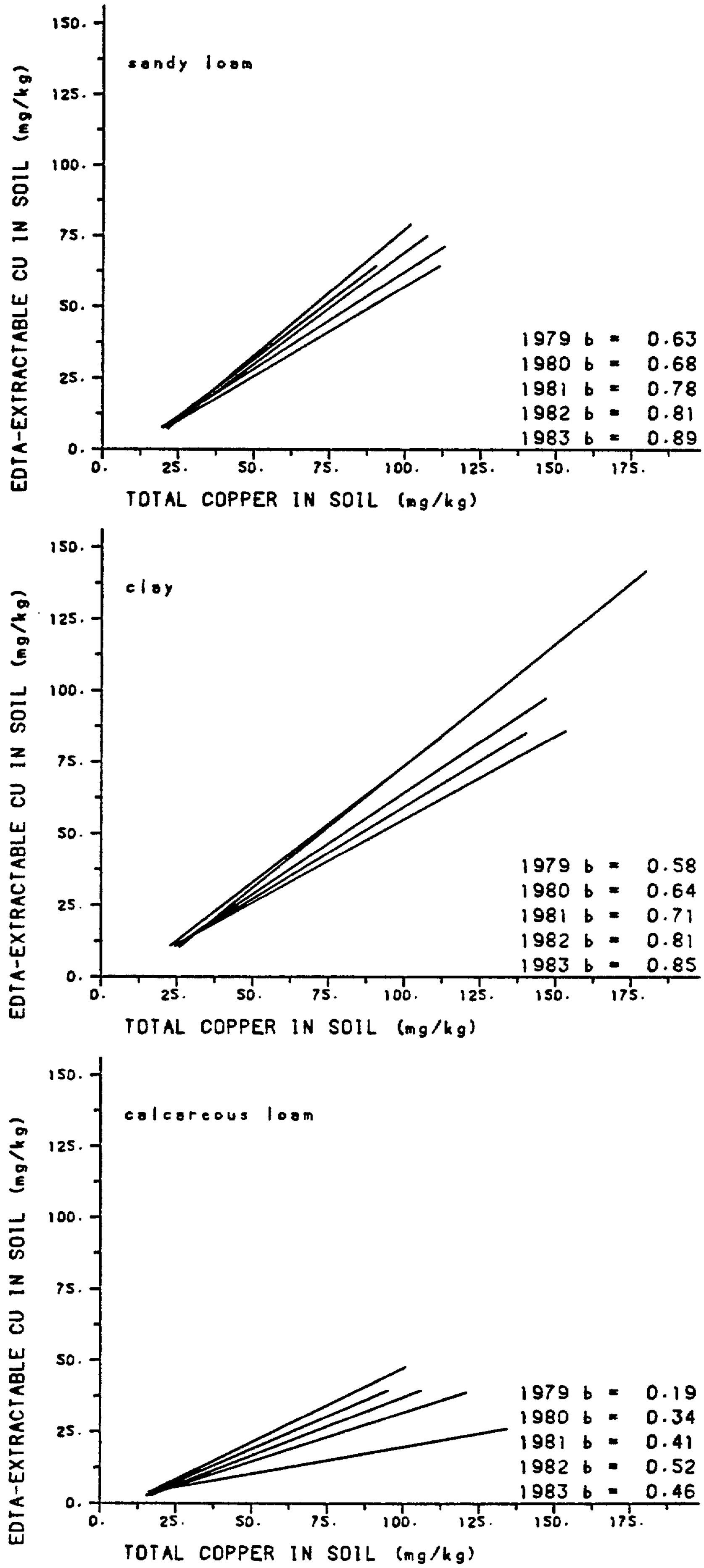


Fig 10. EDTA-extractable Cu in soil vs. total Cu in soil, 1979-83
(liquid sludge treatments)

Estimates of the slope (b) for fitted lines in successive years tend to increase for Cu and judging by standard errors for this estimate the changes appeared significant. This was confirmed by analysis of variance on figures of percent metal extracted by EDTA of total (%EDTA) for all data from liquid sludge treatments. Annual %EDTA data for Cu and other metals together with LSD estimates when the effect was significant are given in Table 10.

Table 10. Mean percent of total metal extracted by EDTA in soil, effects between years for liquid sludge treatments

Metal	Soil	1979	1980	1981	1982	1983	LSD 79/80,80-83
Ni	sl	22	21,22	25	24	27	NA,3.1
	c	25	24,23	25	25	28	NA,2.9
	cl	13	8	7	11	9	1.7
Cu	sl	53	58,58	61	63	67	4.2,3.7
	c	55	59,58	62	68	67	2.6,2.9
	cl	22	29	32	42	36	2.8
Zn	sl	35	36,37	35	36	35	NA,NA
	c	47	43,39	39	38	40	NA,NA
	cl	36	29	29	35	29	4.7
Cd	sl	82	80,84	85	91	94	NA,7.6
	c	90	90,89	89	92	98	NA,3.9
	cl	51	45	49	49	43	4.7
Pb	sl	59	58,62	61	63	66	NA,3.9
	c	66	68,64	64	74	70	NA,3.1
	cl	35	25	27	36	30	2.5

soil type : sl - sandy loam, c - clay, cl - calcareous loam
note : the means for 1979 and the first mean for 1980 on soils sl and c are from soil analyses of Blocks D, E and F only (54 plots/year). The comparison for these by analysis of variance was separate to the comparison for 1980-83 where all 6 Blocks are included (108 plots/year)
LSD : the least significant difference between mean percents is given for each comparison, taken at the 5% level of significance if applicable, otherwise; NA.
treatments excluded : background soil, S1R1 and bed-dried sludge

In order to compare the proportion of metal extracted by EDTA of total in the background soils, the liquid sludges and the bed-dried sludge, mean data was calculated for all five years. Table 11 contains %EDTA means for Ni, Cu, Zn and Zinc

Table 11. Percentage of metal extracted by EDTA of total in soil

Soil	Sludge	n	Ni	Cu	Zn	ZE
sandy loam	none	162	13	37	6	13
	liquid	486	90	77	87	84
	bed-dried	96	63	58	87	69
clay	none	162	13	40	5	13
	liquid	486	94	76	92	87
	bed-dried	96	61	56	87	67
calcareous loam	none	180	4	20	19	12
	liquid	540	20	37	44	35
	bed-dried	96	12	27	26	21

note : for liquid and bed-dried sludge treatments means are based on sludge metal only (background soil metal subtracted see text, equations (5) and (6))

n = number of treatments for original mean concentrations from which these percentages are calculated

Equivalent (ZE). The means were calculated as:

i) for background soil

$$Eo\% = 100 \times SEo/STo \qquad (5)$$

ii) for sludge in soil

$$Es\% = 100 \times (SEs-SEo)/(STs-STo) \qquad (6)$$

where

STo is the mean concentration of total metal in soil (mg/kg)

SEo the mean concentration of EDTA-extractable metal in soil (mg/kg)
STs and SEs total and EDTA mean concentration of metal in sludged soil (mg/kg)
Eo% and Es% percentage of EDTA-extracted metal of total

The %EDTA means for the sandy loam and clay background soils were similar for Ni, Cu, Zn and ZE. The calcareous soil differed for these individual metals but gave a similar ZE figure of 12%. For the liquid sludges much higer %EDTA values are seen, ZE averaging 86% for the non-calcareous soils and 35% for the calcareous loam. Bed-dried sludge metals were extracted by EDTA in proportions somewhere between those in background soil and liquid sludge, except Zn which was similar for both sludge types in the non-calcareous soils.

3.2.2.2
General models for Ni, Cu, Zn and ZE

To estimate equations which would relate EDTA-extractable Ni, Cu, Zn and ZE to total concentrations in each soil type for all liquid sludge treatments, regression analyses were carried out on the same basis as individual years but incorporating data for all five years. In addition because regression coefficients for the sandy loam and clay were similar data was further pooled for these soils to give an equation for 'non-calcareous' soil. The equation which allows estimation of EDTA-extractable metal from total metal and vice versa takes two forms:

i) for sludged soils in this trial and types with comparable background metal concentrations,

$$SE = a + b \times ST \tag{7}$$

ii) for other liquid sludged soils when predicting SE

$$SE = b \times ST \qquad (8)$$

where SE is EDTA-extractable metal concentration in soil (mg/kg)

ST total metal concentration in soil (mg/kg)

a the constant estimate

b the slope estimate

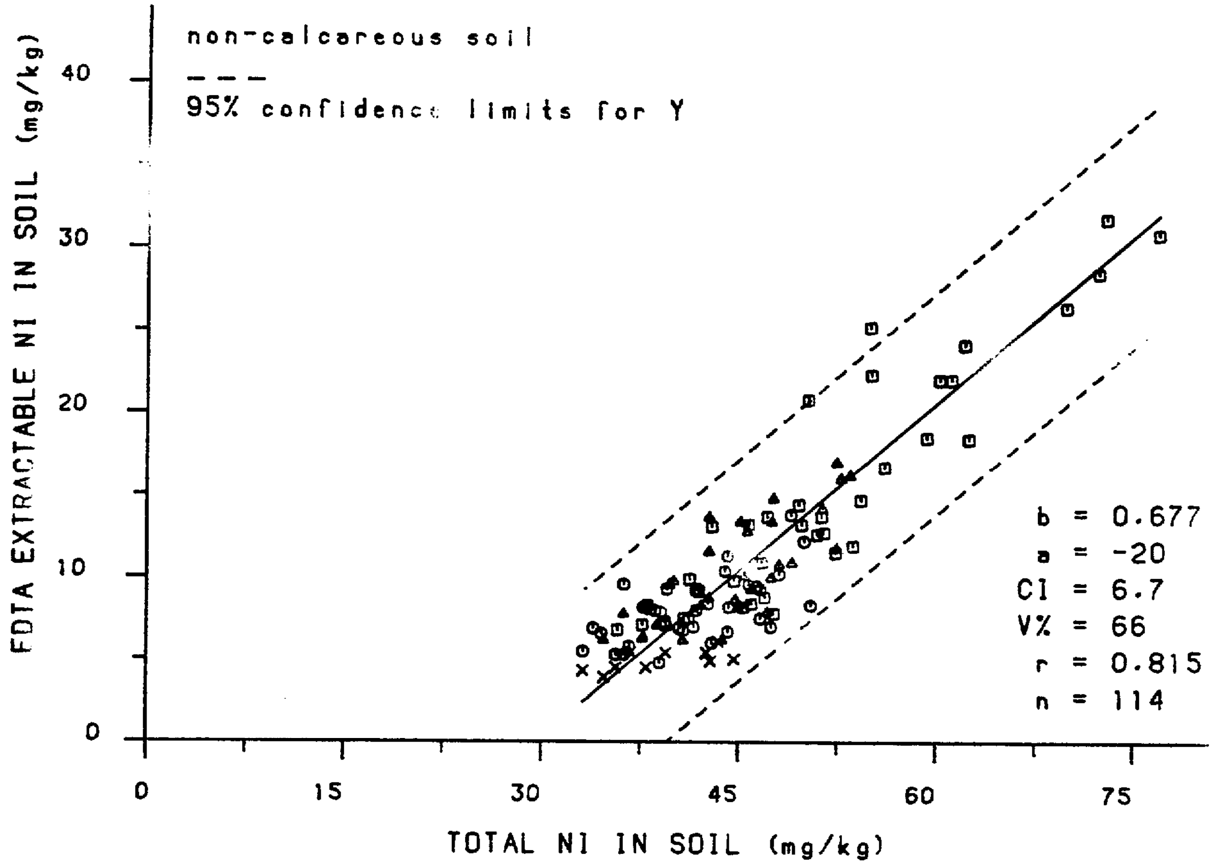
It was shown that %EDTA values in background soil are likely to be lower than that for liquid sludge, see Table 11 Section 3.2.2.1. Therefore, equation (8) gives the best estimate for predicting an EDTA-extractable metal concentration from a total level generally where background soil metal may be less than soils in this trial. It represents the worst possible case, thus ensuring SE is not underestimated. The estimates for b and a together with other statistics, for Ni, Cu, Zn and ZE on each soil type are shown in Table 12. The relationships for a non-calcareous and calcareous loam are illustrated for these metals and ZE in Figures 11-14. The figures show points for annual treatment means, the 'best-fit' straight line based on raw data, and the 95% confidence limits for an individual predicted value of EDTA-extractable metal. The confidence limits were estimated from the initial regression of EDTA metal upon total metal, see Section 3.2.2 and applied to the final line fitted. The confidence interval (CI) is given when $x = \bar{x}$.

Table 12. Estimates of regression coefficients relating total metals to EDTA-extractable metals based on soil analyses for all five years, (background soil and liquid sludge treatments)

Soil	Metal	b(slope)	SE	a(constant)	$\bar{y}$	$\bar{x}$	V%	n
sandy loam	Ni	0.72	0.017	- 20.1	8.9	49.0	63	702
	Cu	0.77	0.009	- 8.4	29.2	40.3	90	702
	Zn	0.87	0.012	- 94.0	56.0	173.0	87	702
	ZE	0.81	0.011	-293.0	186.0	593.0	86	702
clay	Ni	0.77	0.014	- 26.3	11.0	48.4	76	702
	Cu	0.72	0.008	- 6.9	36.1	59.4	92	702
	Zn	0.91	0.009	-116.0	77.0	213.0	93	702
	ZE	0.82	0.008	-351.0	238.0	719.0	93	702
sand and clay (combined)	Ni	0.68	0.011	- 20.0	10.0	44.4	66	1404
	Cu	0.74	0.006	- 7.3	32.7	54.2	91	1404
	Zn	0.87	0.007	-101.0	67.0	193.0	90	1404
	ZE	0.78	0.007	-301.0	212.0	656.0	89	1404
calcareous loam	Ni	0.22	0.005	- 2.3	1.6	17.8	67	776
	Cu	0.37	0.007	- 2.6	14.0	45.1	71	776
	Zn	0.46	0.006	- 19.0	38.0	124.0	87	776
	ZE	0.33	0.004	- 4.1	78.0	357.0	87	774

SE = standard error of the slope estimate b
$\bar{y}$ = mean of EDTA soil metal analyses, liquid sludge treatments

$\bar{x}$ = mean of total soil metal analyses, liquid sludge treatments
V% = percentage variance accounted for by the regression
n = number of raw data points on which regression was based
ZE = zinc equivalent basis



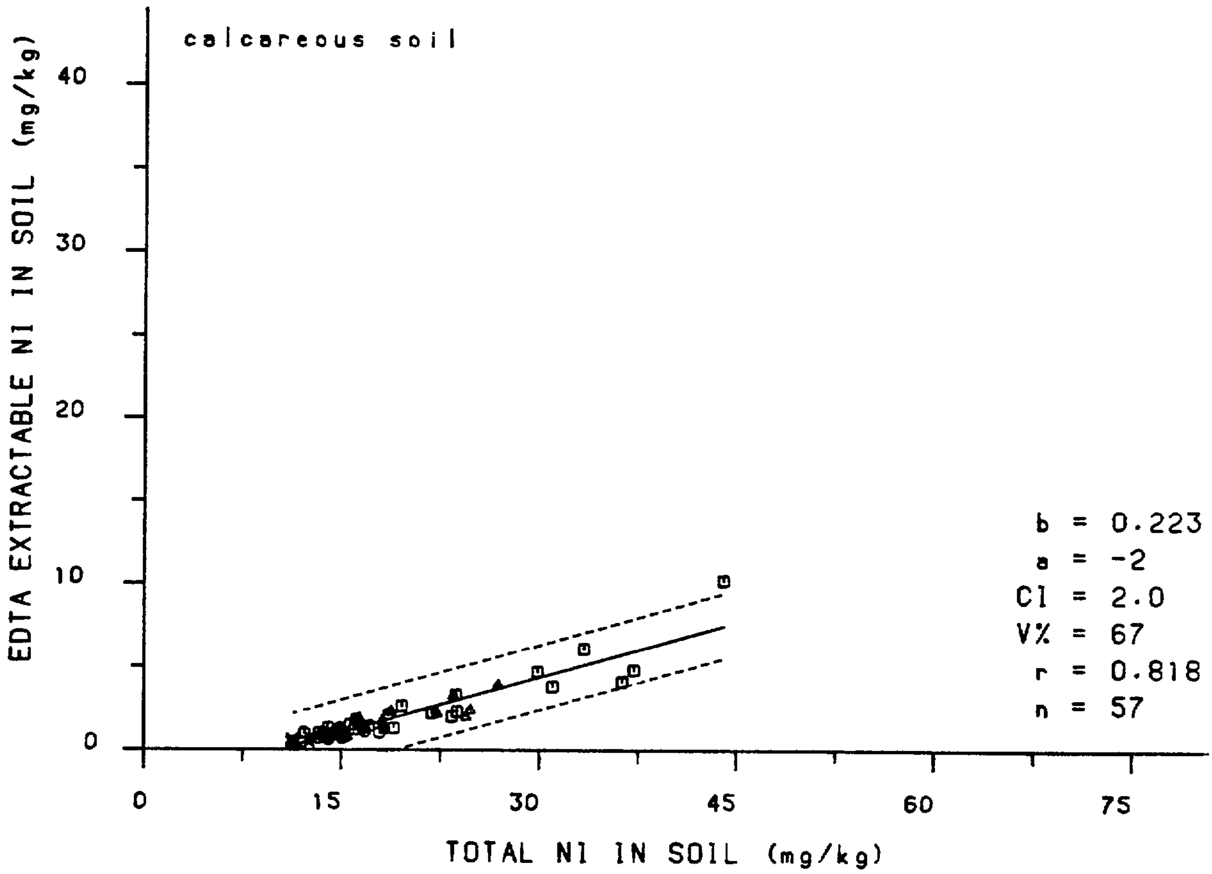
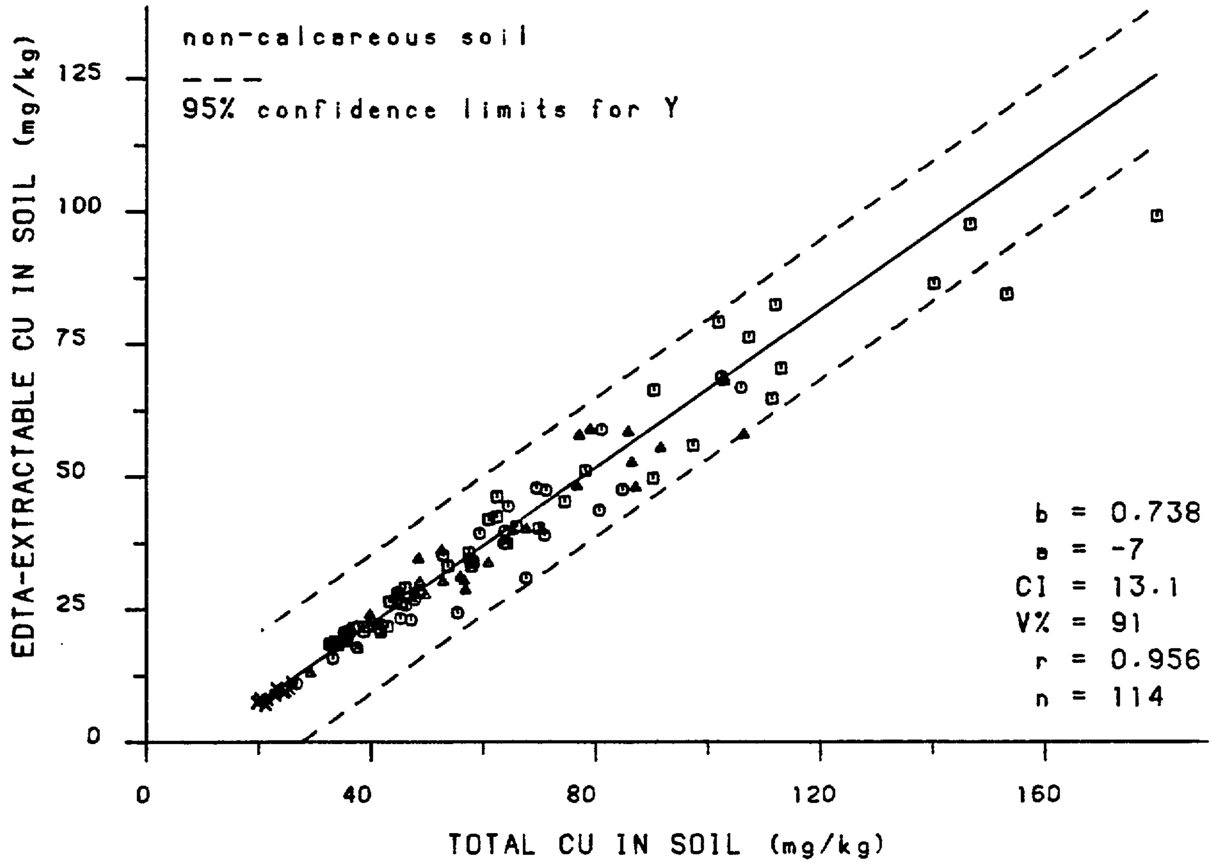


Fig 11. EDTA Ni in soil vs. total Ni in soil.
for non-calcareous and calcareous soil.



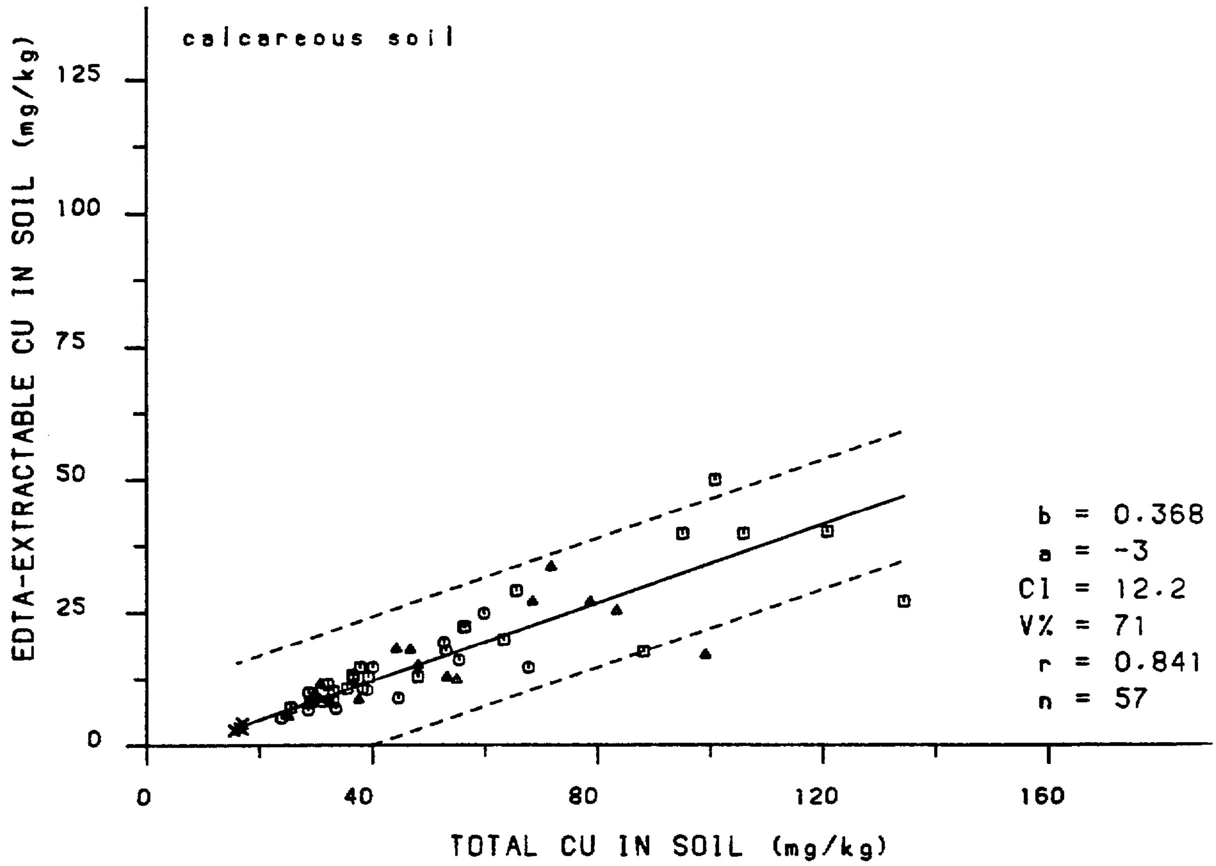


Fig 12. EDTA Cu in soil vs. total Cu in soil. for non-calcareous and calcareous soil.

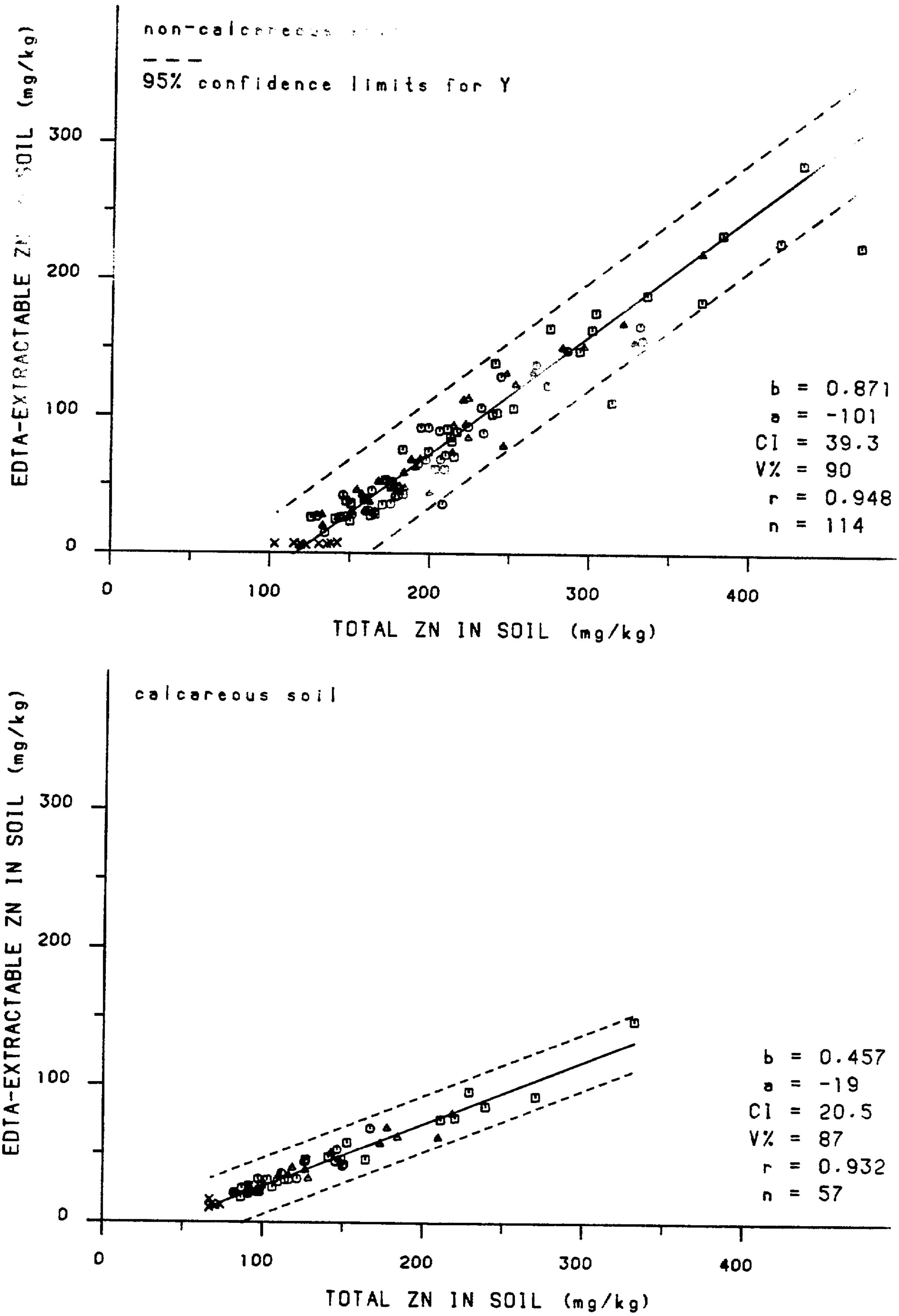


Fig 13. EDTA Zn in soil vs. total Zn in soil.
for non-calcareous and calcareous soil.

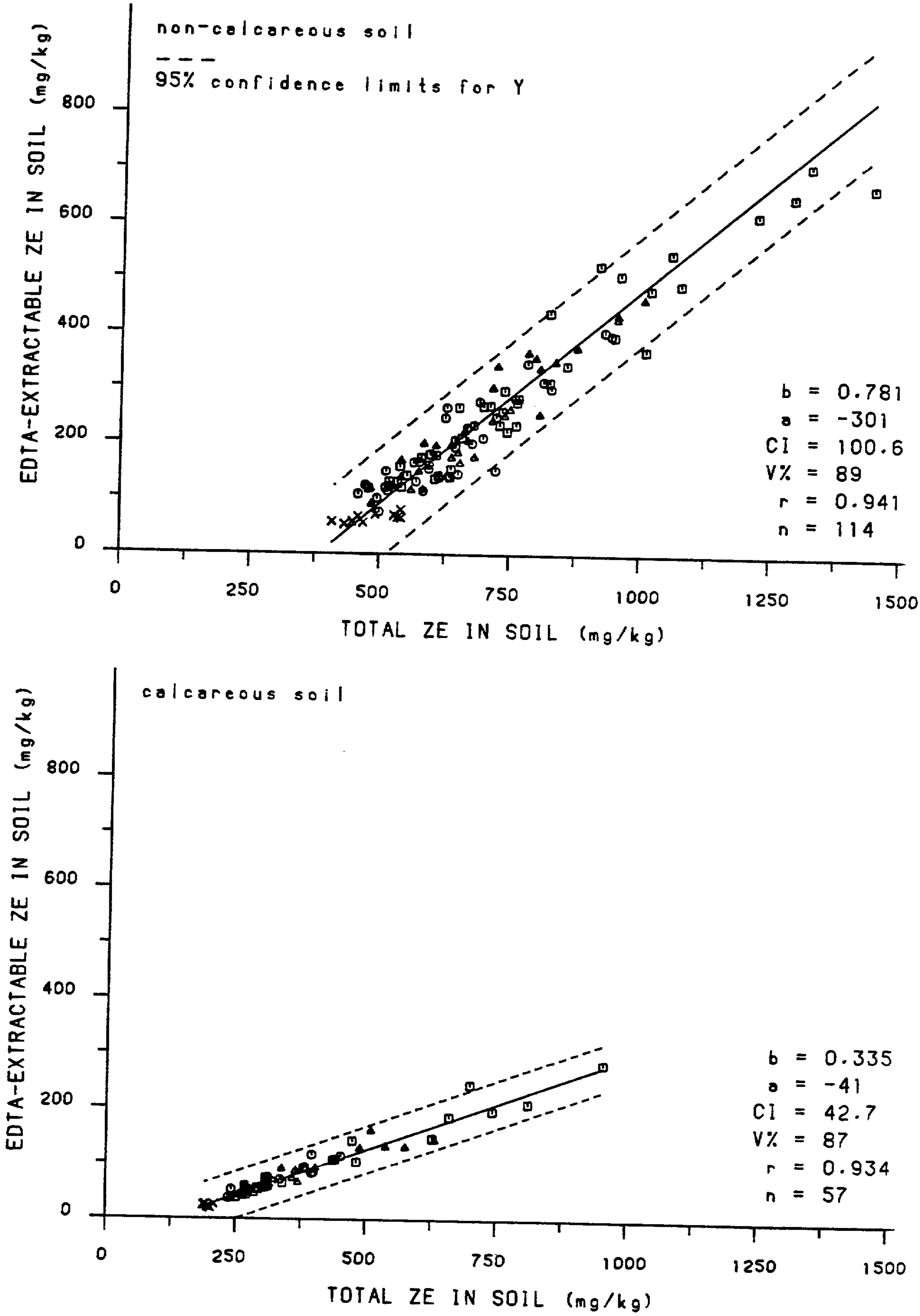


Fig 14. EDTA ZE in soil vs. total ZE in soil. for non-calcareous and calcareous soil.

3.3 Crops yields

The fresh yield of crops was measured for all treatments (Section 2.7) Dry matter (DM) was determined on sub-samples taken for analysis and used to convert fresh yield data to dry yields (tonnes/hectare). Thus, yields were obtained for all six crops grown on the three soil types in five successive years: 90 sets of data, each consisting of 30 treatments. For ryegrass, where two to five cuts were taken each year, total dry yield per year was considered. Mean yields (t/ha DM) for unsludged, liquid sludge, and bed-dried sludge treatments on an annual basis including standard errors (SE) are given in Appendix G.

3.3.1 Effect of sludge application rate on yields

Rates of application of sludge were necessarily much higher than those associated with normal operational practice in order to achieve the desired metal loadings (Tables 3 and 4). The treatment design was such that the mean sludge dry solids addition for each liquid sludge type was similar, but the mean metal level applied was significantly different. This was to enable effects due to sludge nutrients (nitrogen and phosphorus) and conditioning (dry solids) to be separated from the effects of metals added to the soil. Correlation between crop yields and sludge dry solids was measured for all data sets. Correlations between yields and sludge applications are shown in Appendix H. Bed-dried sludge treatments were excluded from these correlations as it was shown that they were different from the liquid sludges. Out of 88 data sets, 36 showed significant effects on yield (Table 13). Of these significant correlations, twenty-three gave positive r values which showed that yields increased as dry solids increased. The remaining

Table 13. Cases where the relation between sludge dry solids addition and crop yield was significant (P<0.05)

Positive correlation

Soil	Ryegrass	Wheat	potato	cabbage	lettuce	red beet
sl	1979,82,83			1981	1979	1979
c		1979	1981	1980,82	1979,81	
cl	1979		1980,81	1979,82,83	1979,81	1979,81,83

Negative correlation

Soil	Ryegrass	Wheat	potato	cabbage	lettuce	red beet
sl		1982,83	1982			
c	1983	1981-83	1982			
cl	1981,82	1981-83				

thirteen correlations (15% of the total) gave negative r values which showed that yields decreased as dry solids increased. These significant effects on yield were attributed to sludge nutrients (primarily nitrogen) and sludge conditioning effects on the soils.

Increases in yield due to dry solids occurred in one quarter of all cases, examples were seen on all soils, for all crops and in all years. Three typical cases are shown (Figure 15). It is suggested that these effects were primarily due to soil conditioning by sludge dry solids, which would result in better water retention in the root zone and ease of root penetration. This was a seasonal effect, most marked on the sandy loam soil and on the calcareous loam which were freely draining. It occurred despite irrigation of crops during droughty conditions in summer.

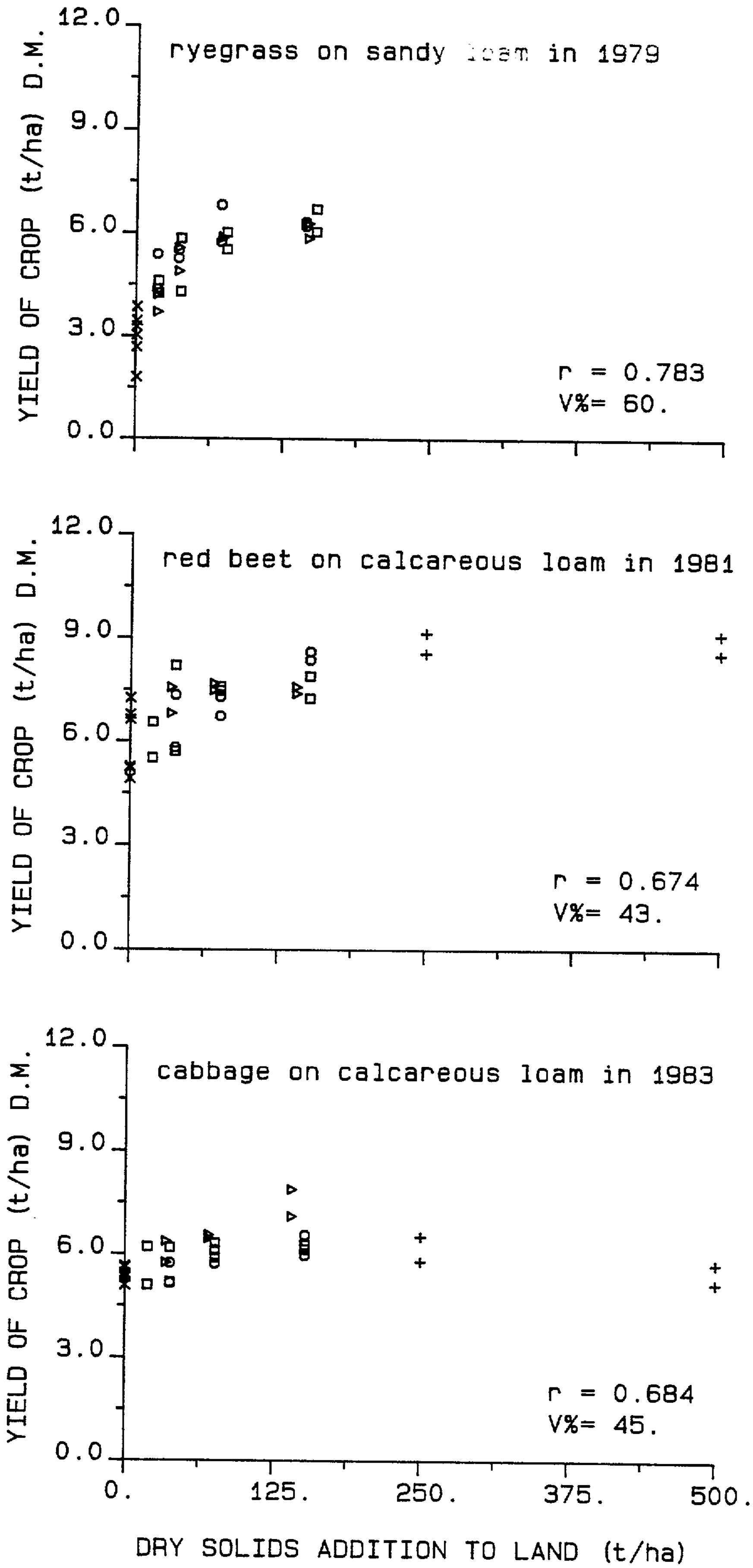


Fig 15. Relationship between yield and dried solids addition, positive correlations.

In the few cases where correlations were significant and negative, the dominant crop was wheat on all three soils 1981-1983. Three cases are shown in Figure 16. Despite application of stem-shortener, lodging of wheat occurred and was recorded in 1980-1982 on the calcareous loam and in 1981-1983 on the non-calcareous soils. The term describes the state of a plant when its stems have bent over and grain yield is reduced as a consequence. Lodging scores covering all treatments on all soils in 1981 are shown in relation to sludge dry solids addition (Figure 17). As above, correlations were found to be significant. The only other cases of decreased yields in relation to sludge dry solids were three ryegrass crops 1981-1983, and two potato crops in 1982. Some lodging scores were recorded for ryegrass: on the non-calcareous soils in 1981 where no significant yield reductions occurred. Judging by the visual appearance of the crops in 1981-1983, it was obvious that growing the leys beyond a normal operational period of three years had led to deterioration of the sward; age and a significant infestation with weed grasses had contributed to reduced yields.

Remaining instances of crop yield reduction could be accounted for by adverse weather conditions. For example, in 1982, when two potato crops suffered reduced yields on the non-calcareous soils, significantly dry weather occurred during the growing season compared with previous years. In contrast in 1983, very wet weather was encountered which led to the worst problems of the trial in connection with establishing marketable crops during the season. The adverse weather forced resowing of potatoes on the clay soil because the initial crop had rotted in waterlogged

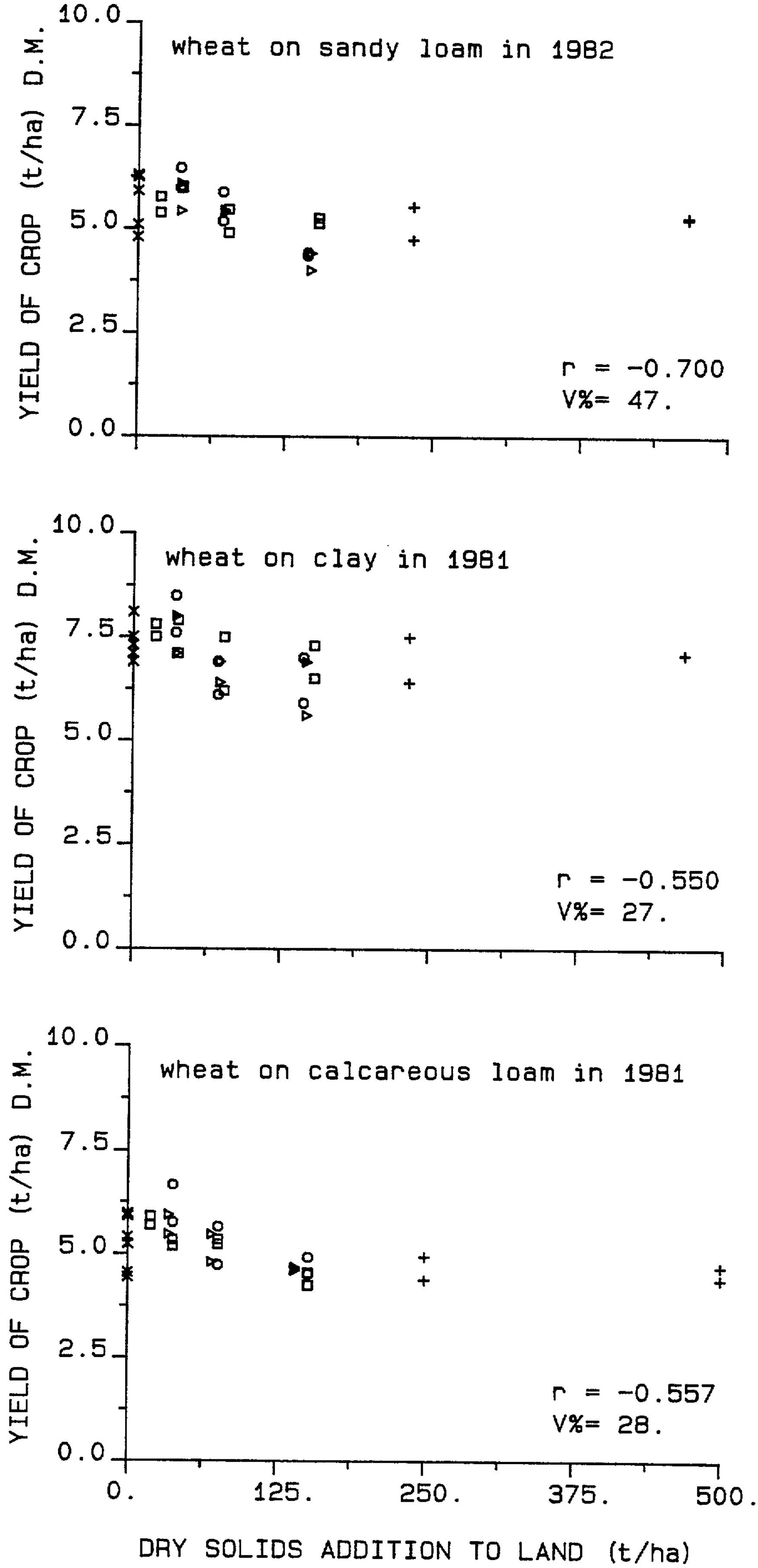


Fig 16. Relationship between yield and dried solids addition, negative correlations.

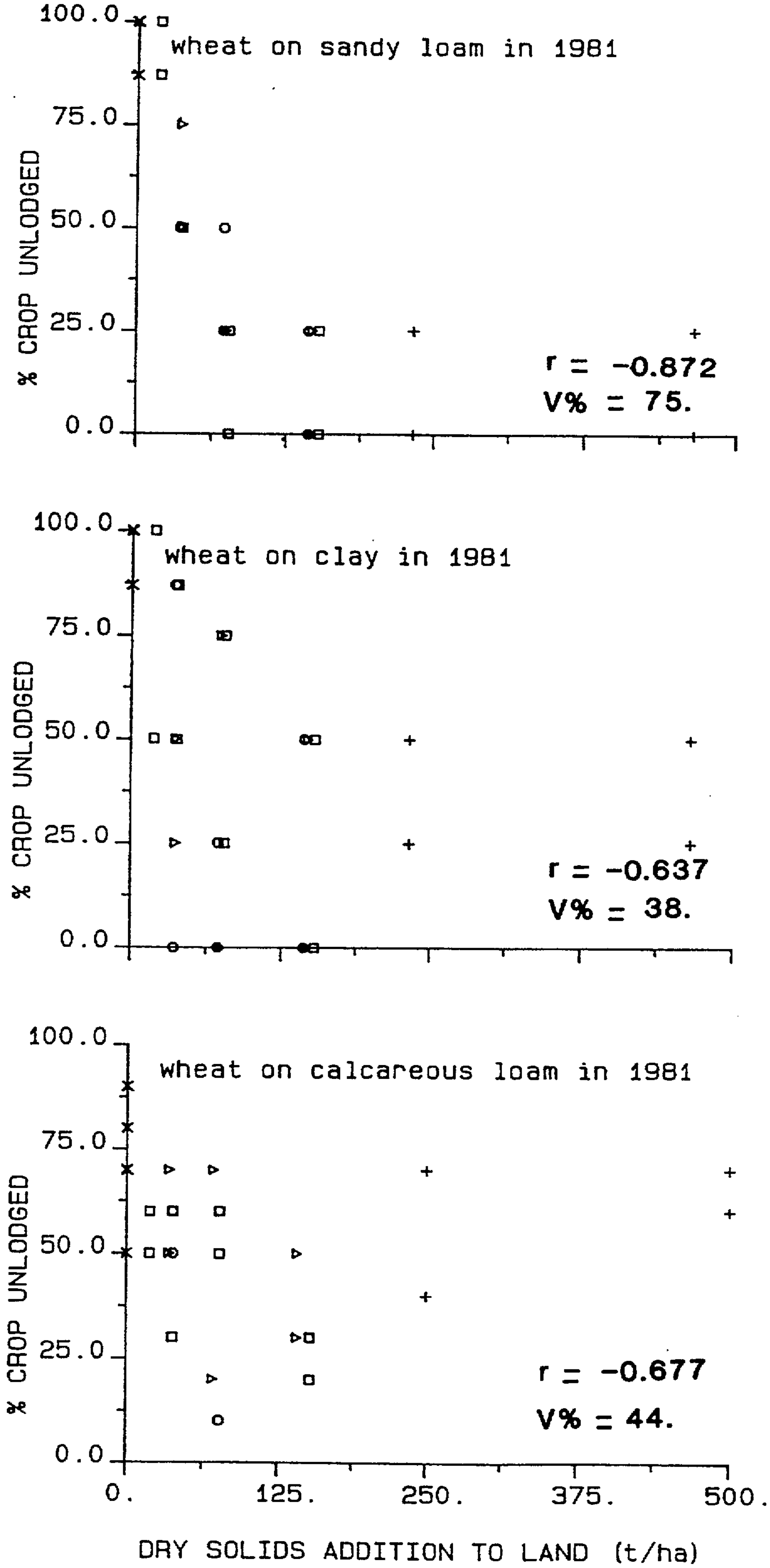


Fig 17. Relationship between wheat lodging and dry solids addition.

ground. Despite all efforts neither the potatoes nor the lettuce were mature when sampled and yield assessment was not possible for these crops on the clay.

The overall effects on yield of sludge dry solids addition to the soils can be summarised by examining mean crop yields for the five year period of the trial. Mean yields for unsludged, liquid sludged, and bed-dried sludged treatments are shown in Table 14. Analysis of variance was used to compare these means. Results showed nine non-significant cases and nine significant effects. Of the significant effects, seven were positive, that is, application of liquid sludge overall had increased yields compared to unsludged treatments. The remaining two negative effects were for wheat on the clay and calcareous loam. These small (6-10%) overall reductions in yield for wheat were thought to be due to lodging in successive years (see above).

3.3.2 Effects of soil metal concentrations on yields

The relationships between yields and sludge metals was determined by analysis of variance for the three liquid sludge types as the factorial experiment was designed specifically for this. Although the mean metal additions to soil for each liquid sludge type were significantly different, mean dry solids were similar for all types of liquid sludge. Mean Ni, Cu, and Zn additions from liquid sludges are given in Table 15. Mean yields from each type of liquid sludge for the five year period are given in Table 16. In the comparison between sludge type, unsludged and bed-dried treatments were separated first. Thus only liquid sludge treatment means were compared (see Equation (1), Section 2.9 (Tkn term)). The analyses of

Table 14. Mean crop yields on unsludged, liquid sludged and bed-dried treatments 1979-1983 (t/ha DM).

Crop	Soil	Sludge none	Sludge liquid	Sludge bed-dried*	Crop	Sludge none	Sludge liquid	Sludge bed-dried
ryegrass	sl	8.0	9.0+	8.6	cabbage	7.3	7.8+	7.6
	c	10.6	10.9	10.8		8.8	9.1+	9.3
	cl	10.4	10.9	11.4		6.4	7.6+	6.9
wheat	sl	5.2	5.9	4.6-	lettuce	1.29	1.38+	1.41
	c	4.9	4.4-	4.6-		1.39	1.54+	1.54
	cl	4.7	4.4-	4.4		0.91	1.17+	1.27+
potato	sl	9.2	9.1	7.6-	red beet	5.9	5.8	6.1
	c	10.3	10.0	10.3		6.6	6.4	7.1
	cl	8.3	9.4	9.3+		6.3	7.6+	8.4+

* Note n = 30 no sludge, n = 90 liquid sludge and n = 16 bed-dried sludge. Bed-dried treatments only cover four years 1980-83.

\+ Means significantly higher than the mean for unsludged plots covering the equivalent period ($P<0.05$).

\- Means significantly lower than the mean for unsludged plots covering the equivalent period ($P<0.05$).

Table 15. Mean metal additions to soil from each liquid sludge (kg/ha)

non-calcareous soils sludge type	Ni	Cu	Zn	ZE
Hogsmill Valley liquid (S2)	7.2	55.0	124	292
Mixed liquid (S1/S2)	11.6	65.0	144	367
Perry Oaks liquid (S1)	24.5	94.7	206	591
calcareous soil:				
S2	5.5	35	97	211
S3 (S1/S2)	10.5	56	123	319
S1	21.0	103	182	556

variance between these means for every crop/soil combination showed that there was no significant effect of liquid sludge type on yield. Further analyses of variance were carried out on an individual year basis for wheat, potato, and ryegrass on all three soils to determine the effect of sludge type on yield in particular years. Once again, no significant effects were found to be present. Thus in every analysis, yield was not affected by the significantly different average loadings of phytotoxic metals in each liquid sludge type.

A third way of assessing the effects of metals on yield is by correlating yield and either theoretical metal additions to soil or actual metal concentrations in soils. As with variance analysis the effects of dry solids on yields cannot be excluded when using this method. Despite this, if reductions in yield were due to metals and not dry solids, for example wheat in 1981, then r values for metal-yield relations should be substantially greater than those for dry solids-yield relations. Correlation coefficients for Ni, Cu, and Zn additions in sludge to soil and total and EDTA-extractable concentrations in soil are shown in Appendix H. Generally, in those cases where significant correlation was demonstrated for dry solids-yield relations, correlations for metal-yield relations were lower. There were a few cases in which yield was shown to be affected by metal concentrations, however, the picture was not at all clear because of other factors related to the nature of sludge on yield. In summary, crop yields were not found to be affected by metals over and above those effects of sludge dry solids.

Table 16. Mean crop yields from each liquid sludge 1979-83 (t/ha DM).

Crop	Soil	Sludge Type S1	S2	S3	SE
ryegrass	sl	9.2	8.9	9.0	0.13
	c	11.0	10.9	10.8	0.22
	cl	10.9	11.1	10.9	0.24
wheat	sl	5.1	4.8	5.1	0.12
	c	4.5	4.4	4.3	0.10
	cl	4.5	4.3	4.4	0.11
potato	sl	9.3	9.0	8.9	0.19
	c	10.4	10.1	9.7	0.28
	cl	9.2	9.4	9.5	0.17
red beet	sl	6.0	5.8	5.6	0.16
	c	6.4	6.6	6.3	0.16
	cl	7.8	7.4	7.7	0.19
cabbage	sl	7.9	7.6	8.0	0.17
	c	9.1	9.3	8.8	0.16
	cl	7.6	7.6	7.7	0.18
lettuce	sl	1.39	1.39	1.38	0.039
	c	1.58	1.54	1.51	0.042
	cl	1.16	1.17	1.16	0.047

SE standard error of means for each crop/soil combination.

Effects of sludge type were not significant ($P>0.05$) in any combination.

n = 30 in all cases except for lettuce and potato on the clay where n = 24 as yields were not determined in 1983.

sl sandy loam, c clay, cl calcareous loam.

3.4 Metals concentrations in crops

Athough Cd, Ni, Cu, Zn, and Pb were determined in the edible portion of all crop samples, in some cases levels of Cd, Ni, and Pb were equal to or less than the detection limit for the analytical method. Metal determinations in crop samples grown on the sandy loam and clay soil to which this applied were reported and entered into all data analysis at the approriate detection limit. Metal results at or below detection limits for these soils are listed in Table F1 of Appendix F. Crop results for the calcareous loam which were most commonly on the detection limit, however, were reported at the calculated 'criteria of detection'. Thus if after blank correction the metal concentration fell below the limit of detection, this absolute value was taken for data analyses. This reduced any positive bias that might otherwise occur in low Cd, Ni and Pb concentrations in the crops for this soil.

Mean concentrations of Cd, Ni, Cu, Zn, and Pb in crops are summarised in Tables I1-I18, Appendix I. Here are detailed means for unsludged, liquid sludged, and bed-dried sludged treatments in each year. Standard errors (SE) for the means of each sludge type are included. The standard errors were estimated from analyses of variance (AoV) on each crop-metal-soil combination. The model for the AoV was based on the basic model (Equation 1, Section 2.9), with an extra source of variation between years included. Bed-dried sludge treatments which did not fit into the full factorial design were dealt with in a separate model.

3.4.1 Relation between metal concentrations in crops and soil

Effects on metal concentrations in crops due to soil parameters were initially studied by examining correlations. For each metal in turn in each year the crop concentration (y-variate) was correlated with six soil parameters: theoretical addition of metal to soil (a), log(a), the measured total metal concentration in soil (c), log(c), EDTA-extractable metal in soil (e) and finally log(e). The correlation coefficients (r values) obtained are listed in Tables J1-J18 of Appendix J. Values of percent variance (V%) accounted for by a simple linear model of y upon x are also given for each case.

Studies of the many scatter plots of crop metal vs. total or EDTA-extractable metal in soil illustrated in earlier reports on this trial, clearly showed that for most cases the liquid sludged treatments followed a similar pattern. Bed-dried sludge treatments, however, just as in the soil relations in Section 3.3 were distinct in that for many cases sludge metals were less available to crops. Consequently bed-dried sludge treatments were excluded from regression analyses including the basic correlations in Appendix J. The relations seen for the remaining unsludged and liquid sludged treatments in many cases approximated to a simple linear model of crop metal estimated by a soil parameter. In some cases, however, points appeared to follow a curvilinear pattern, which approximated to a linear relation after logarithmic transformation (log) of the soil parameter (x-variate). Examples are illustrated in Figure 18 for Cd in wheat in 1981 vs total Cd in soil. The best relation was linear for the sandy loam, but linear-log for the clay and calcareous loam. Thus in this example crop metal (y) could be estimated for the sandy loam as $y = a + bx$ and for the other two soils as $y = a + b.\log(x)$.

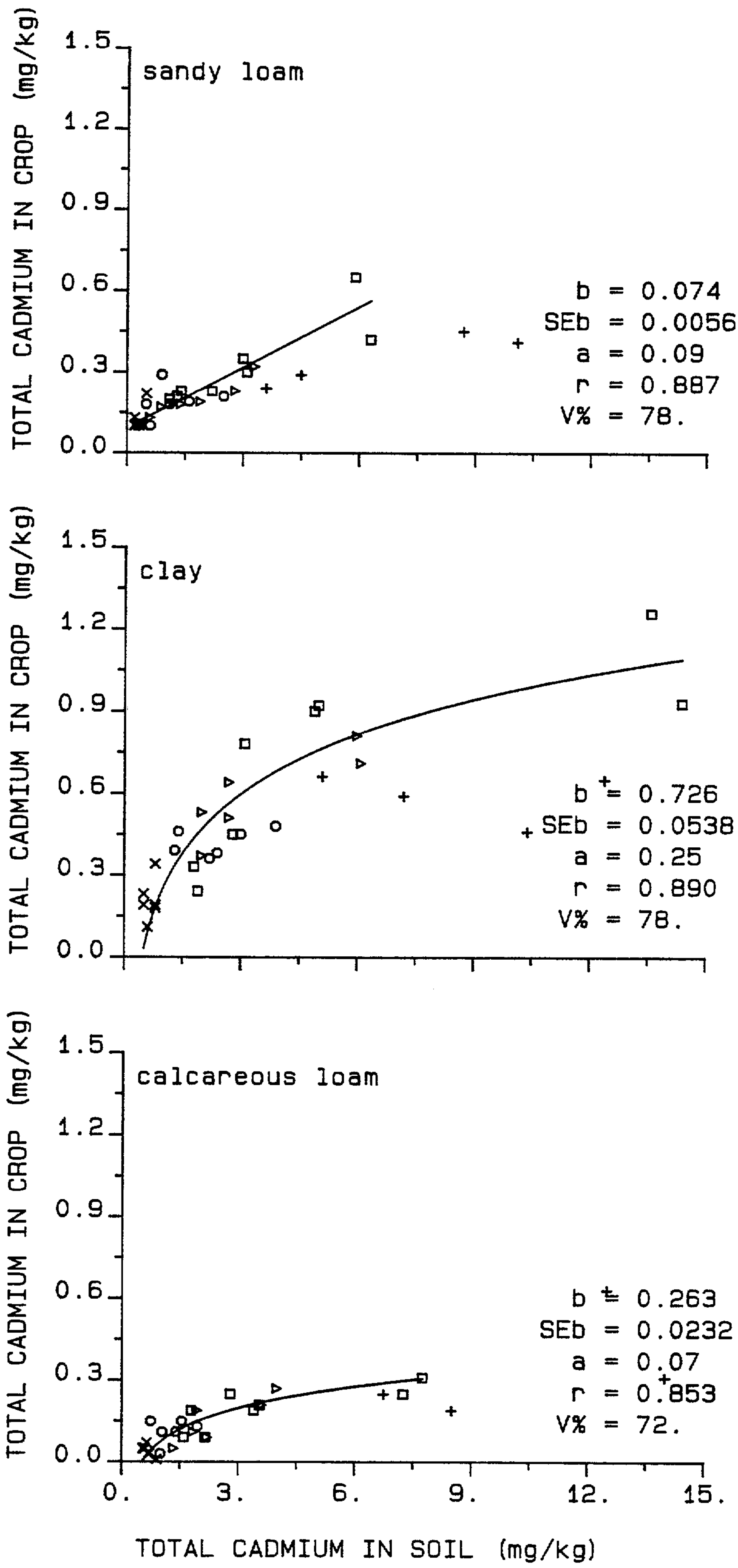


Fig 18. Cadmium in wheat vs. total cadmium in soil, 1981

The regression coefficients, a and b, for each model were not simply estimated from a y upon x regression. Cadmium determinations in soil were not 'independent' variates but subject to significant error in the same order as that associated with Cd levels in the crop so the regression coefficient b was calculated as the geometric mean of b estimates from y upon x, and x upon y regressions, in the same manner as relations between EDTA-extractable metal in soil and total metal in soil (Section 3.2.2). The constant term, a, was then estimated as: a = mean (y) - b.mean (x). In addition the standard error (SE) associated with each b estimate was calculated as

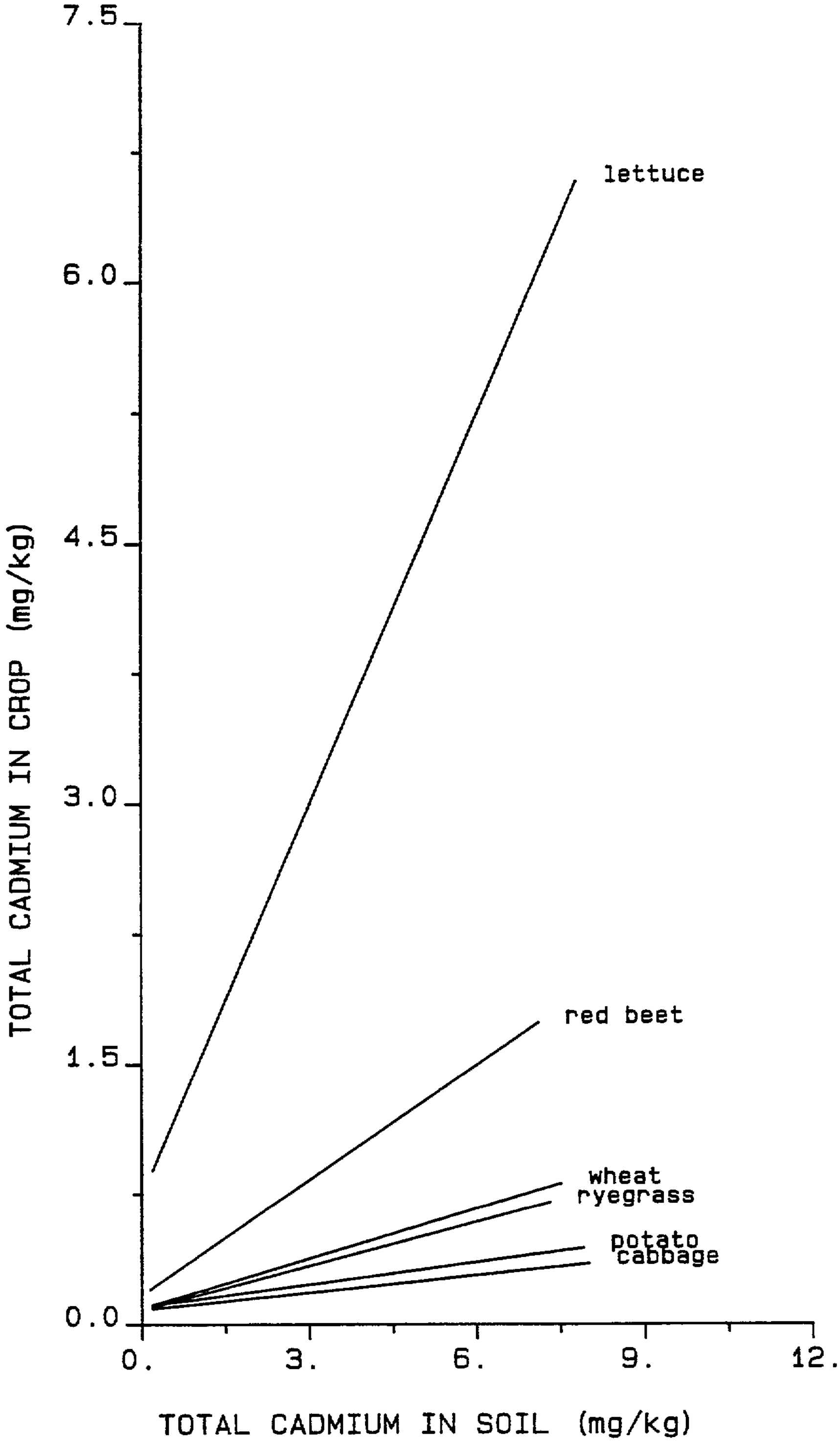
$$SE = b/2 \times \sqrt{[(SE1/b1)^2+(SE2/b2)^2]} \qquad (9)$$

where b1 and SE1 are estimated from a y upon x regression
b2 and SE2 are estimated from a x upon y regression.

Prediction of Cd concentrations in the crops was the main objective of this work and in addition crop-soil relations for one phytotoxic element, Zn, were also studied in detail. Zinc was chosen because measurable uptake occurred in all crops and was frequently significantly correlated to soil Zn. For these metals, it was decided to reconcile the differences in crop-soil relations between soil types and in individual years and adopt the model which fitted the data best overall for each crop. Averaging V% values from correlations in Appendix J it was clear that Cd in wheat grain, for example, was very significantly related to total Cd in soil. This relation was significant in every year on each soil and the mean V% value was 64. Models predicting Cd in crop from EDTA-extractable Cd in soil were similar, and those using Cd addition to soil slightly better. Logarithmic transformation of any soil parameters did not increase V% values overall. Therefore, the model chosen to estimate Cd in wheat throughout the trial was a linear one based on total Cd in soil as no other models were significantly better overall. Thus regression coefficients for models in individual years and also in five years data pooled for each soil were estimated for wheat using this model. This process was carried out for all crops for Cd and Zn. To provide an illustration of the scatter of data, figures have been drawn illustrating all raw data for Cd in crops vs. total Cd in soil (Appendix K). A linear model relating these variates was chosen for all six crops and lines are fitted to data in the figures (excluding bed-dried sludge) and regression statistics given. In contrast Zn in all crops was best described by a linear-log model, estimating crop Zn from log EDTA-extractable Zn (EDTA-Zn) in soil. Figures of raw data, fitted logarithmic curves and statistics for Zn are also included in Appendix K.

3.4.1.1 Wheat

Cadmium in wheat grain was significantly related to total Cd in soil in each season on every soil. Raw data is illustrated in Figures K1-3, Appendix K. Regression statistics for the linear models are contained in Table 17. The gradient (b) of the fitted line for pooled data indicated that the level of Cd in wheat grain grown on liquid sludged non-calcareous soils was one tenth of the total Cd level in the soil plus a constant level (a). For the calcareous soil wheat absorbed Cd at half this rate. The line fitted to data for all five years on the sandy loam soil is illustrated along with lines for other crops in Figure 19. Uptake of Cd



	b	SE
le	0.749	0.0383
rb	0.221	0.0158
wh	0.097	0.0048
ry	0.084	0.0032
po	0.042	0.0041
ca	0.034	0.0043

Fig 19. Cadmium in crops vs. total cadmium in sandy loam 1979-83, unsludged and liquid sludge treatments.

Table 17. Relationship between total Cd in wheat and total Cd in soil, background soil and liquid sludge treatments.

Soil		1979	1980	1981	1982	1983	79-83
sl	b	0.060	0.093	0.074	0.124	0.118	0.097
	SE	0.0055	0.0094	0.0056	0.0077	0.0086	0.0048
	a	0.06	0.14	0.09	0.10	0.09	0.09
c	b	0.057	0.105	0.081	0.090	0.178	0.105
	SE	0.0074	0.0107	0.0075	0.0094	0.0242	0.0074
	a	0.11	0.14	0.23	0.23	0.17	0.16
cl	b	0.023	0.060	0.045	0.057	0.065	0.046
	SE	0.0044	0.0051	0.0046	0.0042	0.0146	0.0031
	a	0.06	0.01	0.03	-0.01	-0.05	0.015

key: 79-83 relation for all five years data pooled
sl sandy loam, c clay, cl calcareous loam
b slope, SE standard error of b, a constant

in wheat grown on bed-dried sludge treatments was significantly lower than for liquid sludge on all

soils. In view of the low uptake and few treatments the relation for this sludge type was not modelled.

Nickel levels in wheat rose little above background levels on any soil and these concentrations were often close to the detection limit of 0.4 - 1 mg/kg, see Appendix F. Despite this, some years gave significant correlations.

Copper in wheat, as for Ni, varied little according to soil Cu. Concentrations in grain grown on bed-dried sludge treatments on the sandy loam in 1980 and 1982 reached a mean maximum level of 10 mg/kg, compared to controls averaging 7 mg/kg.

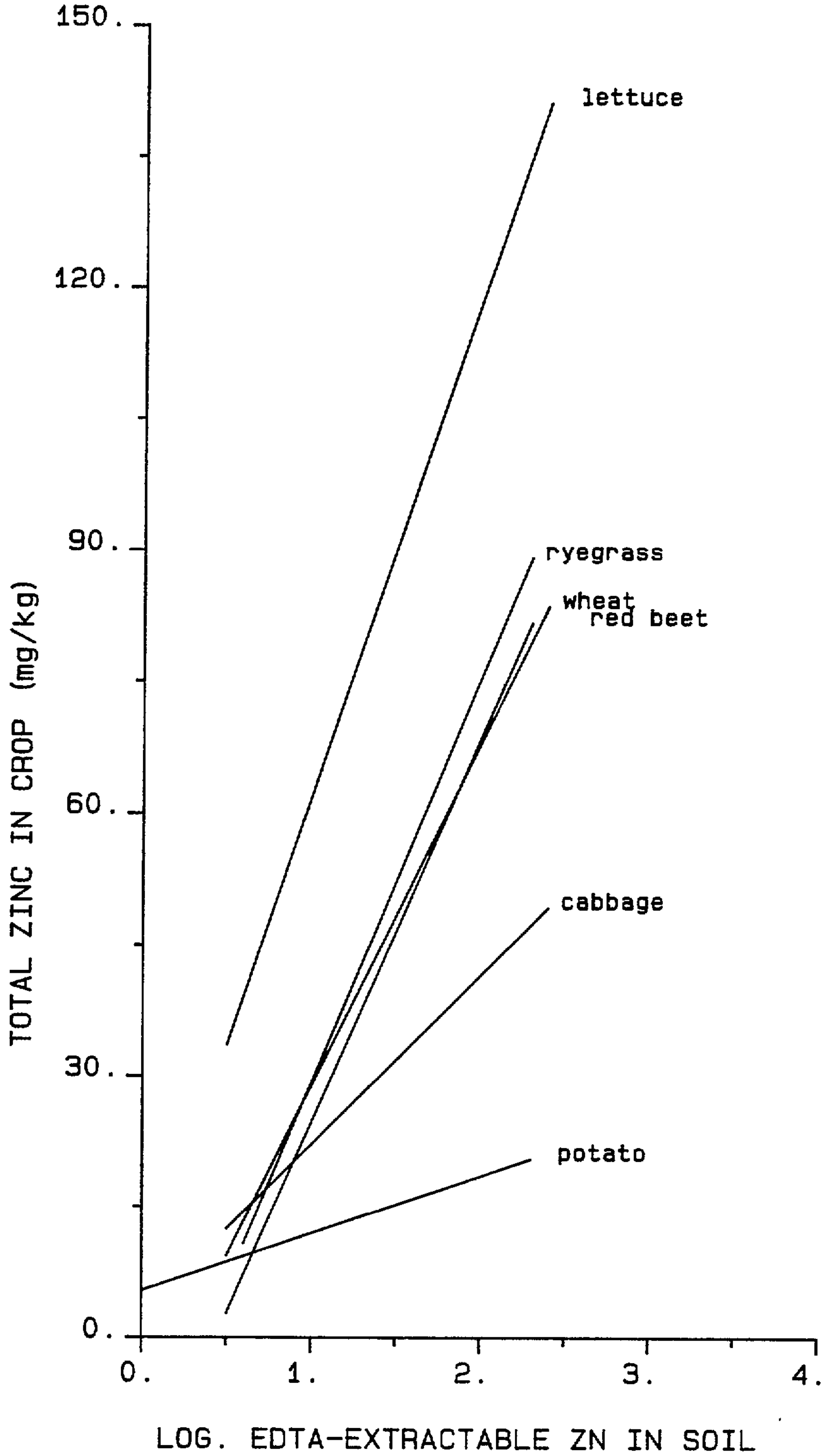
Zinc concentrations in wheat were studied in some detail as with Cd and related best to log.EDTA Zn in soil. Figures K4-6 in Appendix K illustrate the raw data. The regression coefficients are given in Table 18. The regression line fitted to all data, excluding bed-dried treatments, on the sandy loam is illustrated in comparison to those for other crops in Figure 20. The grain Zn levels were broadly similar to the EDTA-extractable levels in soils around the mean values for the liquid sludged treatments.

Table 18. Relationship between total Zn in wheat and total Zn in soil, background soil and liquid sludge treatments.

Soil		1979	1980	1981	1982	1983	79-83
sl	b	29	38	27	41	51	39
	SE	2.8	5.6	1.7	4.3	4.4	2.2
	a	-1	-2	0	0	-28	-9
c	b	13	27	29	24	49	33
	SE	2.7	2.8	1.5	3.0	6.8	2.7
	a	21	3	-7	5	-17	-8
cl	b	11	28	23	41	34	33
	SE	2.3	2.4	2.8	4.0	3.1	4.9
	a	16	22	13	-15	-10	-3

key: 79-83 relation for all five years data pooled
sl sandy loam, c clay, cl calcareous loam
b slope, SE standard error of b, a constant

Lead determinations in wheat were least accurate compared with other metals. This was because there was little apparent uptake above the detection limit of 0.5 - 2 mg/kg and there were no significant effects due to sludge treatment.



	b	SE
le	56.	3.0
ry	45.	1.4
wh	39.	2.2
rb	43.	2.7
ca	20.	1.7
po	7.	0.7

Fig 20. Zinc in crop vs. log. EDTA-Zinc in sandy loam 1979-1983, unsludged and liquid sludge treatments.

3.4.1.2 Potato

Cadmium levels in potato were significantly correlated with total Cd in soil in four out of five years on the sandy loam and calcareous loam soil, but not significant in any year on the clay. The linear relation between total crop and soil Cd had a low gradient compared to other crops except cabbage, see Figure 19. Basic data is illustrated for every year in Figures K7-9, Appendix K. Regression statistics are detailed in Table 19. Where b estimates were significant their standard errors (SE) reflect their limited accuracy. This was because Cd levels in potato grown on the sandy and calcareous loam did not rise greatly above the limit of detection (0.1 mg/kg).

Table 19. Relationship between total Cd in potato and total Cd in soil, background soil and liquid sludge treatments.

Soil		1979	1980	1981	1982	1983	79-83
sl	b	0.046	0.037	0.03*	0.05	0.046	0.042
	SE	0.0076	0.0062	0.012	0.012	0.0084	0.0041
	a	0.08	0.11	0.13	0.11	0.09	0.104
	n	26	26	26	26	26	130
c	correlations were not significant on this soil						
	$\bar{y}$	0.55	0.40	0.48	0.41	0.67	
cl	b	0.020	0.016	0.022	0.034	0.04*	0.028
	SE	0.0036	0.0053	0.0040	0.0041	0.019	0.0030
	a	0.07	0.10	0.02	0.04	-0.01	0.038
	n	26	26	26	26	26	130

* not significant, data included in 79-83 estimate

There were significant correlations between Ni in potato and soil Ni in only two years on the sandy and calcareous loam but in all five on the clay.

The mean concentrations attained in the crop rarely rose above 2 mg/kg. Uptake of Ni from bed-dried sludge treatments was similar to liquid sludge types.

Correlations for Cu were significant in most years but the mean levels in potato tuber grown on unsludged soil of 6-8 mg/kg only increased to 7-13 mg/kg for sludged treatments. Greatest uptake occurred on bed-dried sludge treatments on the calcareous loam.

Zinc levels in potato were low compared to other crops. They are illustrated in relation to EDTA-Zn in soil in Figures K10-12, Appendix K. Many correlations were significant, four years on the sandy loam and clay, and two years on the calcareous loam were modelled. The statistics are shown in Table 20, see also Figure 20. Where the relation was not significant regression coefficients are approximate but included since the technique in estimating them was quite robust (Section 3.4).

Table 20. Relationship between total Zn in potato and total Zn in soil, background soil and liquid sludge treatments.

Soil		1979	1980	1981	1982	1983	79-83
sl	b	8	6.4	5	4.1	3.3	6.6
	SE	1.2	0.73	1.9	0.74	0.71	0.68
	a	5	4	8	9	10	5
c	b	6	4.6	8	6	3*	6.7
	SE	1.8	0.88	1.5	1.6	2.0	0.85
	a	10	11	2	6	13	6
cl	b	14	5*	6	10*	7*	10
	SE	2.5	5.4	1.5	9.4	4.6	2.3
	a	-9	8	8	4	9	1

key: 79-83 relation for all five years data pooled
sl sandy loam, c clay, cl calcareous loam,
b slope, SE standard error of b, a constant
* not significant, results included in 79-83 regression

The vast majority of Pb determinations in potato fell on or below the detection limit of 0.5 - 2 mg/kg. There were no significant correlations between crop and soil Pb concentrations.

3.4.1.3

Lettuce

Raw data for Cd in lettuce related to total Cd in soil is illustrated in Figures K13-15, Appendix K. Correlations were significant in every year on every soil. The line fitted to lettuce Cd in relation to total Cd in soil overall for the sandy loam is included in Figure 19. It had a significantly higher gradient than for other crops. Regression coefficients are summarised in Table 21. On the clay, it proved very difficult to grow a marketable crop, especially during wet seasons. Consequently immature crops were at times sampled rather than lose them. The worst year was 1983 when there was insufficient sample for yield assessment. It is possible that the comparatively high concentrations of Cd found in lettuce from the clay soil were associated with immaturity of the crop. Cd concentrations in bed-dried sludge treatments were lower than for liquid sludge types despite their higher Cd additions.

Nickel concentrations were high in lettuce, the maximum mean value was 10.7 mg/kg for liquid sludge treatments in 1981 on the sandy loam. Correlations, however, were not significant in two and three years on the sandy and calcareous loams respectively. All correlations were significant on the clay.

Although Cu levels were also high in lettuce compared to other crops, significant correlations only occurred in all five years on the sandy loam. Just one year was significant on the calcareous loam and none on the clay.

Table 21. Relationship between total Cd in lettuce and total Cd in soil, background soil and liquid sludge treatments.

Soil		1979	1980	1981	1982	1983	79-83
sl	b	0.46	0.92	0.95	0.92	0.57	0.75
	SE	0.055	0.082	0.084	0.067	0.058	0.038
	a	0.8	0.6	0.6	0.7	1.	0.74
	n	26	25	26	26	26	129
c	b	1.2	1.0	0.7	2.3	6.1*	1.2
	SE	0.14	0.11	0.22	0.36	0.60	0.18
	a	2.7	1.0	3.3	2.2	-3.9	0.24
	n	26	26	26	26	25	104
cl	b	0.26	0.26	0.21	0.28	0.50	0.34
	SE	0.048	0.023	0.026	0.019	0.097	0.025
	a	0.7	0.39	0.44	0.20	0.4	0.37
	n	26	26	26	26	26	130

* very immature samples, excluded from 79-83 regression.

Relations between Zn in lettuce and Zn in soil were strong on the sandy loam and clay but only significant in the first two years on the calcareous loam. Regression coefficients for models estimating Zn in lettuce from log.EDTA-Zn in soil are given in Table 22. Data is illustrated in Figures K16-18, Appendix K.

Mean Pb values were around the detection limit in lettuce grown on the sandy loam and clay, and no significant correlations with soil Pb occurred. There was one exception to this in 1983 on the clay but the crop had been affected by adverse weather, mentioned earlier in this section. On the calcareous loam mean Pb concentrations in the crop were somewhat higher but no significant correlations occurred with soil parameters in any year.

Table 22. Relationship between total Zn in lettuce and total Zn in soil, background soil and liquid sludge treatments.

Soil		1979	1980	1981	1982	1983	79-83
sl	b	53	52	43	61	41	56
	SE	5.1	5.2	3.8	5.6	2.8	3.0
	a	21	17	40	-12	11	7
c	b	97	50	40	63	187	100
	SE	20.3	6.3	3.4	7.4	27.4	9.7
	a	-36	5	32	23	-158	-47
cl	b	36	40	20*	35*	36*	38
	SE	4.6	4.8	17.1	14.0	32.5	3.3
	a	14	8	49	13	24	12

key: 79-83 relation for all five years data pooled
sl sandy loam, c clay, cl calcareous loam
b slope, SE standard error of b, a constant
* not significant, data included in 79-83 estimate

3.4.1.4

Red beet

Cadmium in red beet was significantly related to all Cd soil parameters on all soils. Mean levels were higher than other crops except lettuce. Raw data illustrated in Figures K19-21, Appendix K, show many cases of lower Cd levels in red beet grown on bed-dried sludge compared to liquid sludge treatments. Regression statistics are in Table 23 and the line fitted to all data for the sandy loam is included in Figure 19.

Nickel correlated significantly with total Ni in soil in two, four, and one year on the sandy, clay, and calcareous soils respectively. Mean Ni concentrations in the crop ranged from the detection limit of 0.5 mg/kg on the calcareous loam

Table 23. Relationship between total Cd in red beet and total Cd in soil, background soil and liquid sludge treatments.

Soil		1979	1980	1981	1982	1983	79-83
sl	b	0.29	0.17	0.143	0.23	0.11	0.22
	SE	0.038	0.015	0.0066	0.029	0.036	0.016
	a	0.34	0.20	0.17	0.17	0.23	0.17
	n	26	26	26	26	26	130
c	b	0.24	0.29	0.19	0.26	0.13	0.24
	SE	0.033	0.045	0.053	0.047	0.016	0.021
	a	0.7	0.7	0.6	0.4	0.56	0.52
	n	26	26	26	26	26	130
cl	b	0.08	0.05	0.083	0.082	0.075	0.072
	SE	0.012	0.010	0.0066	0.0070	0.0088	0.0048
	a	0.05	0.11	0.10	0.00	0.02	0.06
	n	26	26	25	26	26	129

to 6.8 for liquid sludged treatments in 1979 on the sandy loam.

Copper concentrations in red beet did not increase far above the mean value for unsludged treatments of 7.9 mg/kg for all soils. The crop levels correlated significantly with total Cu in soil in two years on the sandy and calcareous loams and in none on the clay.

Zinc concentrations in red beet in relation to log.EDTA-Zn in soil are illustrated in Figures K22-24 in Appendix K. Correlations for this relation were significant in every case and regression coefficients are given in Table 24. Mean levels in the crop were similar to those in wheat and ryegrass and this is reflected in fitted lines for the sandy loam (Figure 20).

Table 24. Relationship between total Zn in red beet and total Zn in soil, background soil and liquid sludge treatments.

Soil		1979	1980	1981	1982	1983	79-83
sl	b	39	56	40	50	20	43
	SE	7.9	6.0	2.2	7.3	2.4	2.7
	a	-3	-41	-16	-26	9	-18
c	b	36	72	48	42	37	55
	SE	3.0	9.1	5.7	3.8	2.7	3.7
	a	2	-32	-18	-13	-14	-29
cl	b	13	16	17	17	14	18
	SE	1.6	2.1	2.0	2.5	1.4	1.3
	a	14	9	6	3	5	3

key: 79-83 relation for all five years data pooled
sl sandy loam, c clay, cl calcareous loam
b slope, SE standard error of b, a constant

Lead determinations in red beet were close to detection limits on all soils. They did not show any significant correlation with soil Pb.

3.4.1.5 Cabbage

Levels of Cd in cabbage leaves were generally lower than in any other crop, particularly on the sandy loam and clay. The regression line fitted to all unsludged and liquid sludged data relating crop Cd to total Cd in sandy loam soil had a low gradient, see Figure 19. The raw data on a yearly basis is illustrated in Figures K25-27, Appendix K. The regression coefficients are summarised in Table 25. Significant relations are seen in most years. Cadmium in cabbage grown on bed-dried sludge treatments as a proportion of total Cd in soil was similar to that for liquid sludge in most cases.

Table 25. Relationship between total Cd in cabbage and total Cd in soil, background soil and liquid sludge treatments.

Soil		1979	1980	1981	1982	1983	79-83
sl	b	+	0.031	0.019	0.03	0.06	0.034
	SE	-	0.0044	0.0015	0.011	0.012	0.0043
	a	-	0.1	0.086	0.10	0.09	0.082
	n	26	26	26	26	18	122
c	b	0.04*	0.032	0.037	0.06	0.03*	0.051
	SE	0.061	0.0060	0.0074	0.014	0.011	0.0083
	a	0.22	0.29	0.18	0.10	0.12	0.15
	n	26	26	26	26	26	130
cl	b	0.044	0.036	0.027	0.03*	0.07	0.044
	SE	0.0036	0.0028	0.0059	0.028	0.016	0.0033
	a	0.09	0.05	0.04	0.05	-0.06	0.02
	n	26	26	25	26	26	129

* not significant, data included in 79-83 estimate
+ Cd in crop samples on limit of detection (0.1 mg/kg)

Nickel concentrations in cabbage grown on unsludged treatments averaged 3.3 mg/kg for all soils and increases above this due to liquid sludge treatments correlated significantly in two years on the sandy loam and in all five on the clay and calcareous loam. The maximum mean concentration in the crop of 5.8 mg/kg occurred for bed-dried sludge treatments in 1980 on the clay.

Levels of Cu in cabbage were low compared to other crops with little difference between soil types, however, significant correlations occurred in all cases except 1983 in the sandy loam. Maximum mean concentrations were found for bed-dried sludge treatments.

The relations between Zn in cabbage and log.EDTA-Zn in soil are illustrated in Figures K28-30, Appendix K. Correlation in all cases was significant and regression coefficients estimated are given in Table 26. Mean Zn concentrations in cabbage were low compared to other crops except potato. The crop-soil relation illustrated in Figure 20 indicates significantly higher uptake of Zn by cabbage compared to potato but lower than the other crops.

Table 26. Relationship between total Zn in cabbage and total Zn in soil, background soil and liquid sludge treatments.

Soil		1979	1980	1981	1982	1983	79-83
sl	b	15	18	12	21	15	20
	SE	2.2	1.6	1.2	1.4	5.6	1.7
	a	5	13	9	-1	18	3
c	b	17	26	20	22	17	23
	SE	1.9	1.3	2.0	2.6	1.7	1.2
	a	9	1	2	-3	3	-1
cl	b	28	22	27	10	18	25
	SE	2.5	1.6	3.3	2.6	2.5	2.4
	a	-5	8	-13	10	8	-4

key: 79-83 relation for all five years data pooled
sl sandy loam, c clay, cl calcareous loam
b slope, SE standard error of b, a constant

Lead determinations, as in most other crops, were at detection limits. For example, two-thirds of all results for Pb in cabbage grown on the non-calcareous soils were equal to or less than the detection limit, see Appendix F. There was only one case where correlation was significant between crop Pb and soil Pb in 1979 on the sandy loam, however, the relation was not strong, the mean Pb level in cabbage for liquid sludge treatments was only 1.2 mg/kg in this case.

3.4.1.6 Ryegrass

Total Cd levels in ryegrass and soil are illustrated for every treatment in Figures K31-33, Appendix K. Concentrations in the crop were in the range 0.1 - 1 mg/kg. Correlations were very significant in all cases reflecting the quality of data obtained with this crop. Regression coefficients for linear models relating Cd in ryegrass to total Cd in soil are given in Table 27. The line fitted to all data for the sandy loam soil is included in Figure 19 and is similar to that for wheat. Cadmium in ryegrass grown on bed-dried sludge treatments, excluded from regression analyses, was generally lower in relation to Cd in soil than liquid sludge treatments.

Table 27. Relationship between total Cd in ryegrass and total Cd in soil, background soil and liquid sludge treatments.

Soil		1979	1980	1981	1982	1983	79-83
sl	b	0.098	0.089	0.069	0.093	0.073	0.084
	SE	0.0090	0.0109	0.0056	0.0049	0.0031	0.0032
	a	0.08	0.08	0.10	0.10	0.07	0.09
	n	24	26	26	26	26	128
c	b	0.098	0.074	0.086	0.060	0.054	0.079
	SE	0.0108	0.0171	0.0105	0.0070	0.0043	0.0051
	a	0.22	0.20	0.23	0.17	0.16	0.19
	n	26	26	26	26	26	130
cl	b	0.022	0.030	0.061	0.036	0.016	0.040
	SE	0.0020	0.0035	0.0037	0.0024	0.0017	0.0034
	a	0.017	0.05	0.03	0.07	-0.014	0.014
	n	26	26	26	26	26	130

Correlations for Ni in ryegrass and soil Ni parameters as with Cd were consistently very significant. Mean concentrations ranged from 1.1 mg/kg for unsludged treatments to 21.4 for bed-dried sludge treatments on the calcareous loam. This maximum mean value was, however, untypical because ryegrass on bed-dried sludge and two unsludged treatments was resown late in 1980 after application of this sludge on all soils (Section 2.3). Metal concentrations were higher in later slower growing cuts of ryegrass. Overall, Ni levels were higher in ryegrass than for any other crop.

Copper concentrations in ryegrass increased significantly in relation to all Cu measures in soil in all cases. Mean values exceeding 20 mg/kg occurred but for untypical bed-dried sludge treatments in 1980, see above. Mean crop concentrations ranged from 6 - 15 mg/kg for other treatments.

The data for Zn in ryegrass in relation to log.EDTA-Zn in soil is illustrated in Figures K34-36, Appendix K. In every case correlations were significant. Regression coefficients for the straight line equations describing this relation are summarised in Table 28. Mean concentrations in the crop ranged from 22 - 91 mg/kg.

Lead concentrations in ryegrass were generally higher than in other crops. Particularly high values were found in cuts taken after the first in each year and although later cuts were low yielding the yearly weighted means still reflect this factor. There were just three cases where correlation between Pb in ryegrass and soil Pb were significant, but only just and occurred in the

Table 28. Relationship between total Zn in ryegrass and total Zn in soil, background soil and liquid sludge treatments.

Soil		1979	1980	1981	1982	1983	79-83
sl	b	41	56	41	42	43	45
	SE	3.1	3.9	1.9	2.0	1.9	1.4
	a	-4	-25	-13	-12	-18	-16
c	b	30	37	41	42	38	38
	SE	2.5	5.9	3.1	2.6	2.3	1.8
	a	4	10	-8	-15	-12	-5
cl	b	30	28	36	27	21	31
	SE	1.7	1.7	1.8	1.5	1.6	1.3
	a	8	14	2	11	11	6

key: 79-83 relation for all five years data pooled
sl sandy loam, c clay, cl calcareous loam
b slope, SE standard error of b, a constant

first and final years of the trial, times when the ryegrass was least well established.

3.4.2 Differences between soils

Comparisons of the amount of metal available for plant uptake between soil types can be made over the five year period by examining regression coefficients for lines fitted to all liquid sludge data (crop concentrations versus soil concentrations of metals) for this period. These values were given on an individual crop basis in Tables 17-28, values for Cd in all crops are summarised in Table 29. The constant value, a, represents the Cd concentration in the crop due to background total Cd in unsludged soil and should remain approximately constant between years. The gradient of the line, b, represents the fraction of the total Cd concentration in liquid sludged soil that is found in the crop tissue dry matter, within the range of soil concentrations achieved. Fitted lines are illustrated for Cd in wheat, ryegrass, and red beet in Figure 21, crops for which the relation was significant in every season. The pattern for these crops was similar, little difference between the non-calcareous soils, but the gradients for the calcareous loam were significantly lower. From Table 29 it is seen that for lettuce, which had the highest tissue metal levels, a significant difference also occurred between the gradient for the sandy loam and clay. For potato and cabbage which had the lowest Cd tissue concentrations the small differences in the models for each soil were not significant.

Table 29. Regression coefficients for linear equations describing relation between Cd in crop and total Cd in soil, unsludged and liquid sludge treated soils.

		sandy loam	clay	calcareous loam
wheat	b	0.097	0.105	0.046
	a	0.09	0.16	0.02
potato	b	0.042	NS	0.028
	a	0.10	NS	0.04
lettuce	b	0.75	1.2	0.34
	a	0.74	2.4	0.37
red beet	b	0.22	0.24	0.072
	a	0.17	0.52	0.06
cabbage	b	0.034	0.051	0.044
	a	0.08	0.15	0.02
ryegrass	b	0.084	0.079	0.040
	a	0.09	0.19	0.01

note: equations are $y=a+b.x$ and are fitted to all unsludged and liquid sludge treatment data 1979-83, $n<=130$
where y is the estimated total Cd in crop (mg/kg DM)
and x is total Cd in soil (mg/kg air-dry)

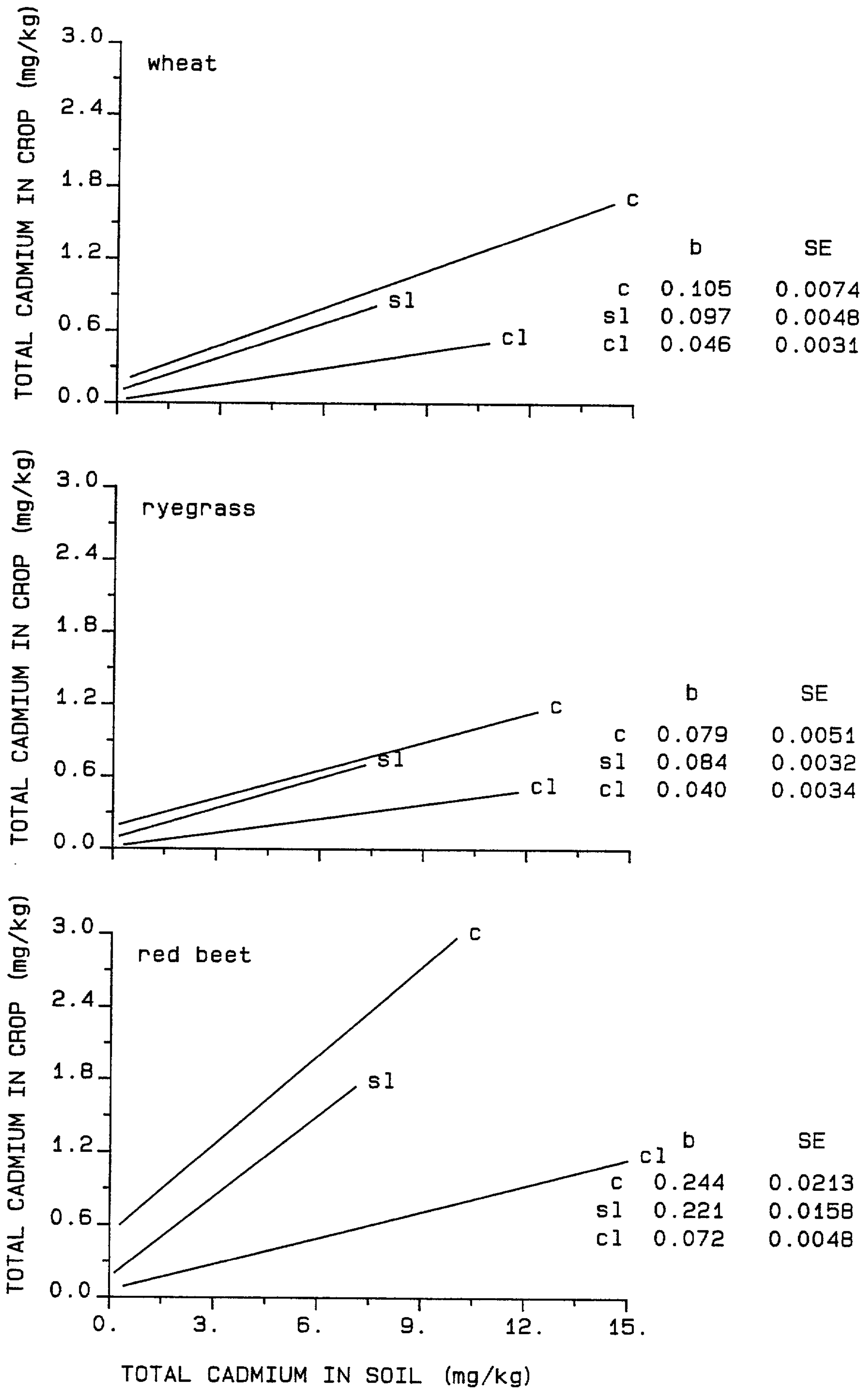


Fig 21. Cadmium in crop vs. total cadmium in soil 1979-83, unsludged and liquid sludge treatments.

In comparison, there was no significant difference between soils for Zn relations in wheat and ryegrass (Figure 22). The distinction between the non-calcareous and calcareous soil in red beet Zn levels was similar to that seen for Cd.

3.4.3 Effects of time

Differences in Cd and Zn crop-soil relations between years were significant. The gradient estimates (b) for lines fitted to individual years for each crop were given along with their standard errors in Tables 17-28. Considering wheat and ryegrass, Tables 17 and 27, for which fitted lines are also illustrated in Figures 23 and 24, many examples of seasonal variation can be seen. It was judged that differences between b estimates for each year for a particular metal-crop-soil case were significant when they exceeded four times the average SE for all years. A broken line fitted to all data for each crop-soil combination is also included in Figures 23 and 24 labelled P. It was important to establish whether this seasonal variation followed an overall trend of the gradient (b) reducing or increasing with time.

On all three soils in 1979 spring wheat was sown, see Section 2.4, and therefore results for this variety might be expected to be different to winter wheat sown in subsequent years: previous work has shown different Cd concentrations in different wheat cultivars grown under the same environmental conditions and in the same sludge treated soil (Davis and Carlton-Smith 1980). Gradient estimates for 1980-83 show no definite trend for any soil although on the clay the value for the final year is significantly higher than for 1980.

Ryegrass data was more accurate than for wheat but the chemistry of metal availability with time might

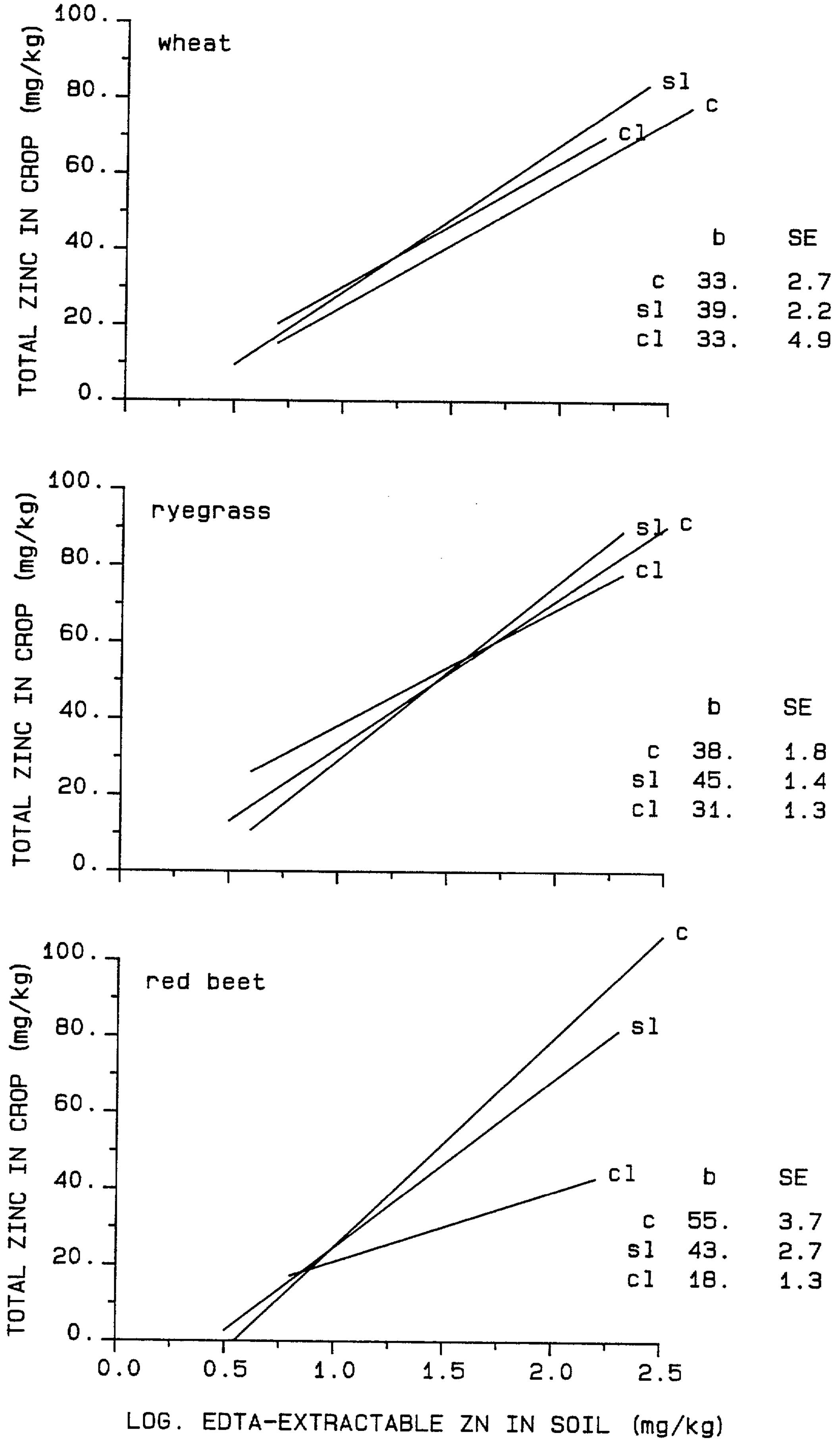
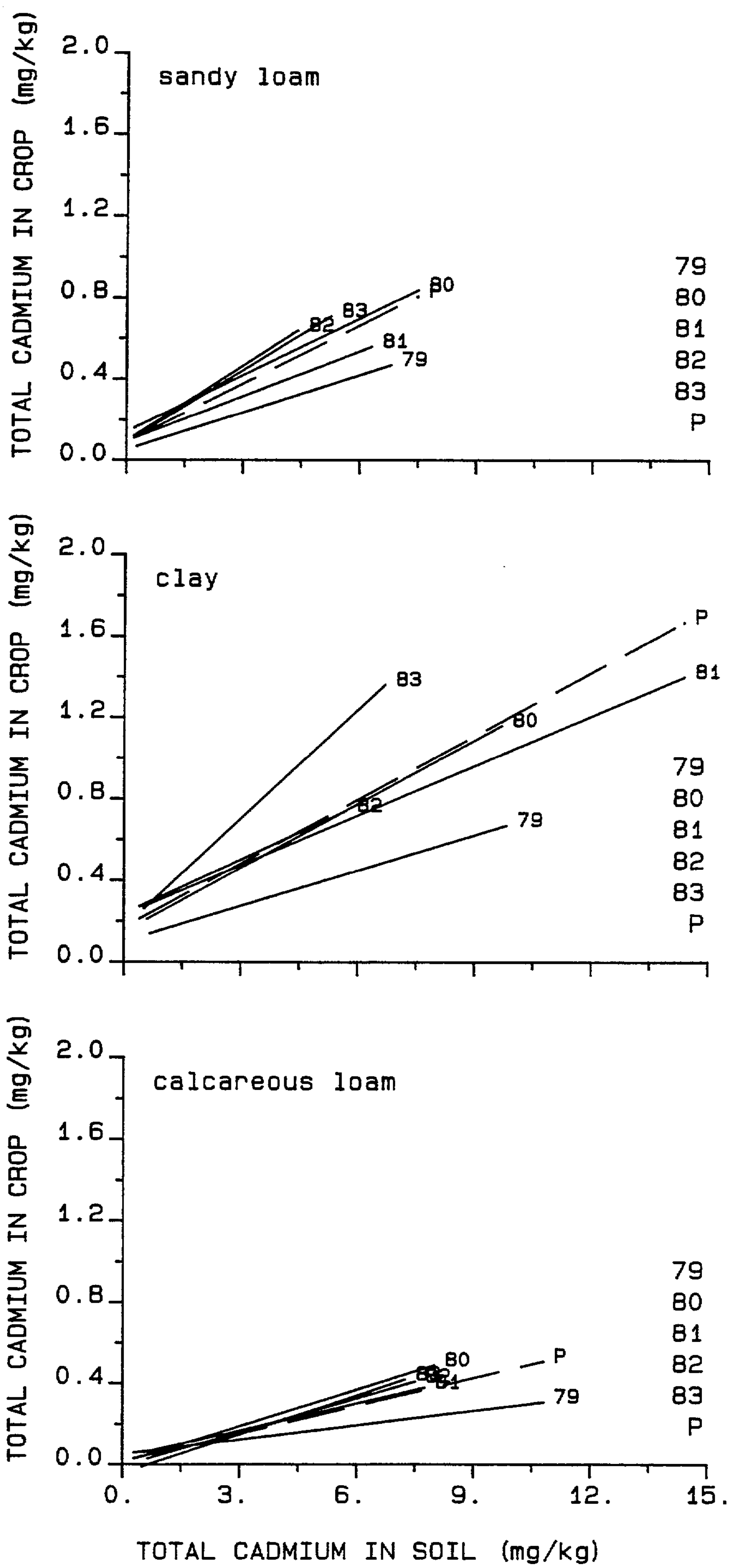


Fig 22. Zinc in crop vs. log EDTA-Zn in soil 1979-1983 unsludged and liquid sludge treatments.



Sandy loam

	b	SE
79	0.060	0.0055
80	0.093	0.0094
81	0.074	0.0056
82	0.124	0.0077
83	0.118	0.0086
P	0.097	0.0048

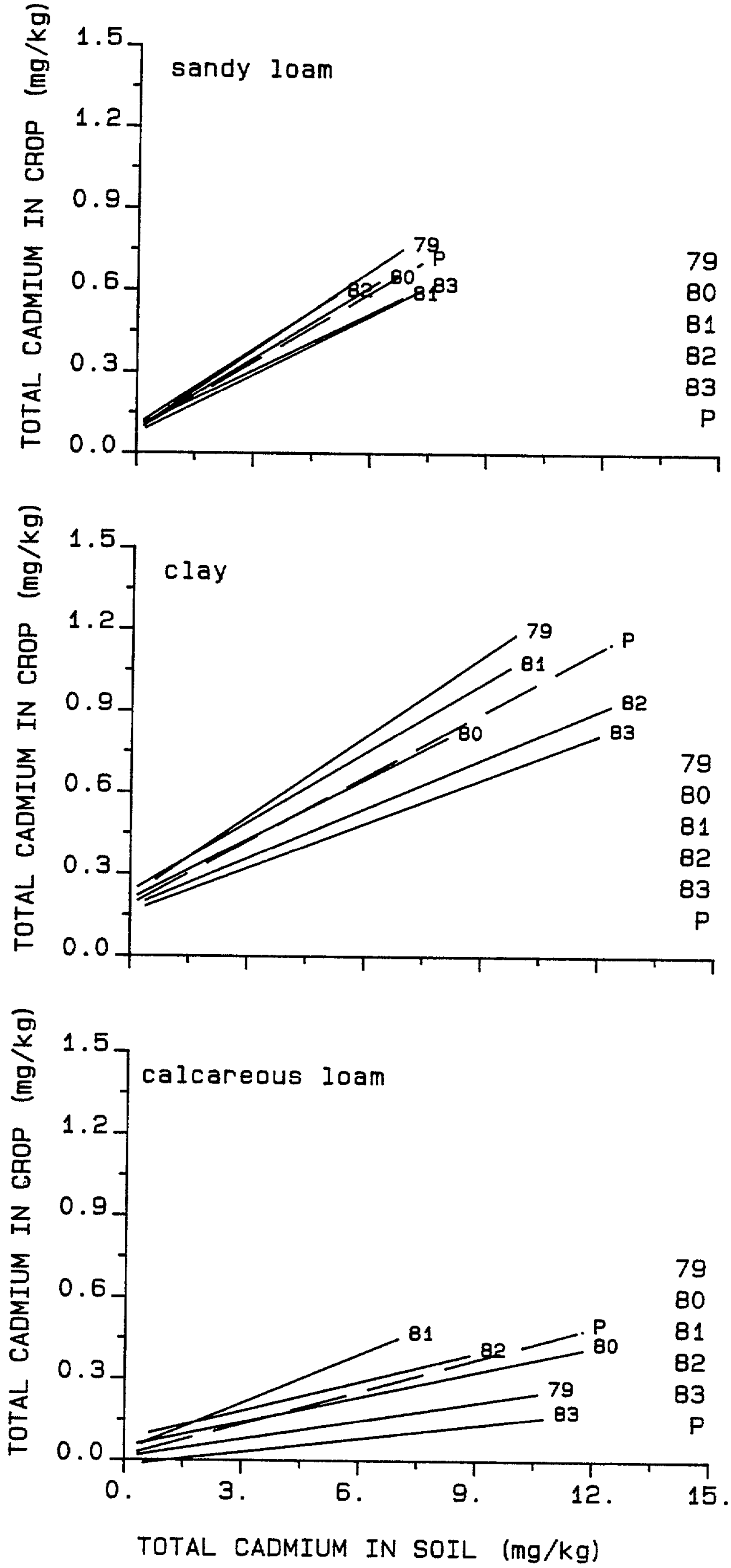
Clay

	b	SE
79	0.057	0.0074
80	0.105	0.0107
81	0.081	0.0075
82	0.090	0.0094
83	0.178	0.0242
P	0.105	0.0074

Calcareous loam

	b	SE
79	0.023	0.0044
80	0.060	0.0051
81	0.045	0.0046
82	0.057	0.0042
83	0.065	0.0146
P	0.046	0.0031

Fig 23. Cadmium in wheat vs. total cadmium in soil 1979-83, unsludged and liquid sludge treatments.



	b	SE
79	0.098	0.0090
80	0.089	0.0109
81	0.069	0.0056
82	0.093	0.0049
83	0.073	0.0031
P	0.084	0.0032

	b	SE
79	0.098	0.0108
80	0.074	0.0171
81	0.086	0.0105
82	0.060	0.0070
83	0.054	0.0043
P	0.079	0.0051

	b	SE
79	0.022	0.0020
80	0.030	0.0035
81	0.061	0.0037
82	0.036	0.0024
83	0.016	0.0017
P	0.040	0.0034

Fig 24. Cadmium in ryegrass vs. total cadmium in soil 1979-83, unsludged and liquid sludge treatments.

be expected to differ because soil was not cultivated each season as with the other arable crops. Clearer trends are seen for ryegrass on all soils, Figure 24 and Table 27, which suggest a decrease in Cd availability to ryegrass with time from the liquid sludge treated soils, however, this effect was less definite on the calcareous loam.

3.5 Crop uptake of metals

The quantity of metal taken up by a crop from soil can be estimated by the product of crop yield and metal concentration in the tissue. Using data from all years, mean uptake of Cd, Ni, Cu, Zn, and Pb was calculated per year in background soil and each sludge type for ryegrass leaves, the heaviest yielding crop of this trial. The increase in metal uptake for sludged treatments above the background soil level should represent metal uptake of sludge origin. Table 30 contains mean background levels and increases for ryegrass, all figures are grammes of metal removed by ryegrass leaves per hectare per year.

Means range from 0.5 g/ha/yr of Cd from the calcareous loam, to 512, of Zn from the clay. Amounts of Cd and Pb removed by sludge treatments are lowest, those for Ni and Cu are approximately ten times higher and amounts for Zn one hundred times higher. Differences between sludge types broadly reflect the different mean sludge additions of metal to soil. Mean sludge additions for Ni, Cu, and Zn were given earlier in Table 15, Section 3.3.2, those for Cd and Pb are in Table 31. Mean increases in metal uptake by each sludge expressed as percentages of the mean additions, provide a measure of the proportion of sludge metal removed by the crop each year. The percentages for ryegrass are given in Table 32. They highlight the small fraction of total sludge metal in soil that is taken up by the crop each year.

Table 30. Ryegrass uptake of metals, means for background soil and mean increases in uptake for each sludge 1979-83 (g/ha/yr).

Metal	Soil	Background soil	Sludge increases S1	S2	S3	S4*
Cd	sl	0.82	2.84	1.01	1.54	1.12
	c	1.91	4.02	1.97	3.63	2.95
	cl	0.52	1.68	0.50	0.76	3.35
Ni	sl	24.5	71.9	23.5	39.5	22.1
	c	39.9	66.0	23.7	45.6	35.1
	cl	15.6	44.0	7.9	17.2	136.
Cu	sl	49	54	57	57	56
	c	86	38	44	48	46
	cl	96	40	32	32	71
Zn	sl	189	470	388	408	237
	c	300	431	476	512	290
	cl	360	268	204	212	392
Pb	sl	13.7	4.9	4.8	4.8	1.4
	c	23.8	1.2	2.1	2.9	1.4
	cl	34.3	4.7	4.5	3.4	1.0

* yearly mean increase in metal uptake 1980-83
n = 30 for all means except S4 where n = 16

Table 31. Mean Cd and Pb additions to soil by each sludge (kg/ha)

Metal	Soil	Sludge type S1	S2	S3	S4
Cd	sl and c	6.1	1.8	2.9	13.2
	cl	6.1	1.1	2.7	15.8
Pb	sl and c	73	56	60	161
	cl	44	38	39	157

Table 32. Yearly mean increases in metal uptake by ryegrass as a percentage of sludge metal additions and background fractions.

Metal	Soil	Sludge type S1	S2	S3	S4	Background fraction*
Cd	sl	0.047	0.056	0.053	0.0085	0.61
	c	0.066	0.109	0.125	0.022	0.41
	cl	0.028	0.045	0.028	0.021	0.11
Ni	sl	0.29	0.33	0.34	0.028	0.16
	c	0.27	0.33	0.39	0.045	0.14
	cl	0.21	0.14	0.16	0.16	0.29
Cu	sl	0.057	0.104	0.088	0.025	0.52
	c	0.040	0.080	0.074	0.021	0.53
	cl	0.039	0.091	0.057	0.025	0.87
Zn	sl	0.23	0.31	0.28	0.055	0.38
	c	0.21	0.38	0.36	0.068	0.33
	cl	0.15	0.21	0.17	0.078	0.78
Pb	sl	0.0067	0.0086	0.0080	0.0009	0.11
	c	0.0016	0.0038	0.0048	0.0009	0.11
	cl	0.011	0.012	0.0087	0.0006	0.09

* Background fraction equals mean uptake on unsludged plots as a fraction of the equivalent background soil addition.

4. DISCUSSION

4.1 Total concentrations of metals in soils

Total metal concentrations in this trial have shown the importance of measuring soil density and depth of sludge incorporation into the soil in order to estimate increases in total metal in soil that will occur as a result of sludge additions.

Theoretically the rise in soil metal concentration (Mi) resulting from sludge addition could be estimated as: $Mi=10 \times Ml/(\rho xs)$ mg/kg where Ml was the metal loading rate (kg/ha), ρ the soil density

(g/cm^3), and s was the depth of incorporation of sludge (cm) (see Equation 3, Section 3.2.1). For most treatments there was good agreement between estimates based on this formula and measured soil concentrations made 5 years after sludge was applied. At this time, lower than expected soil concentrations of Cu and Zn were measured in liquid sludge treatments on the calcareous loam, and of all metals in bed-dried sludge treatments on all soils. The probable explanation for this lies in movement of sludge-treated soil out of the theoretical sampling zone of the plot concerned. This could occur either by lateral movement beyond the plot boundary or by incorporation beyond 15 cm depth; either effect could result from repeated cultivations.

4.2 Extractable concentrations of metals in soil

All soils were extracted with EDTA in this trial (MAFF/ADAS 1978) as limits for Ni, Cu, and Zn in guidelines for sludge disposal to agricultural land in the UK are based on this extractant (DoE/NWC 1981). In addition it has been shown that Ni concentrations in soil can be determined with greatest accuracy by this method (Carlton-Smith and Davis 1983).

The percent of total metal extracted by EDTA from the soil did not vary significantly between the liquid sludge types for any one metal. It varied widely, however, between background soil, liquid and bed-dried sludge treatments and between soil types.

EDTA-extractable Ni, Cu, Zn and zinc equivalent (ZE) expressed as a percentage of total metal in unsludged soils were lower than for metals of sludge origin:

	unsludged soil			
	Ni	Cu	Zn	ZE
non-calcareous	13	39	6	13
calcareous	4	20	19	12

For metals of sludge origin the percentages of total which were extractable by EDTA were:

	Liquid sludges				Bed-dried sludges			
	Ni	Cu	Zn	ZE	Ni	Cu	Zn	ZE
non-calcareous	68	74	87	78	62	57	87	88
calcareous	22	37	46	33	12	27	26	21

To decide to what extent these differences reflect the amount of metal removed by ryegrass, mean values for Ni, Cu, and Zn in liquid and bed-dried sludge are summarised in Table 33.

Although there was some correlation between E and U values according to soil type for Ni and Zn in liquid sludge, the measures of EDTA-extractable metals appear to have little significance to the amount of these metals taken up by ryegrass.

Table 33. Comparison between EDTA-extractable proportion of sludge metal in soil (E) and proportion of total metal taken up by ryegrass per year (U).

Sludge	Soil	Ni		Cu		Zn	
		E	U	E	U	E	U
liquid	sl	90	0.32	77	0.083	87	0.27
	c	94	0.33	76	0.065	92	0.32
	cl	20	0.17	37	0.062	44	0.18
bed-dried	sl	63	0.028	58	0.025	87	0.055
	c	61	0.045	56	0.021	87	0.068
	cl	12	0.16	27	0.025	26	0.078

However, crop concentrations of the most plant-available metals (Cd, Zn and Ni) generally followed the pattern of EDTA-extractability being highest on the non-calcareous soils and liquid sludge treatments. Other mild extractants used on sludged soils to indicate metal availability to plants have proved to be more successful than EDTA. Data are available for several others on certain soil samples in this trial (Appendix E).

4.3 Crop yields

A problem encountered in experiments with sludge occurs in the separation of beneficial effects of the plant nutrients and organic matter it contains from the potentially harmful effects of metals: this applies particularly to yield data. The effects on soil physical condition (Hall and Coker 1981) and nutrient effects (Hall et al 1983) have been reported for the liquid sludge type used in this trial. In the design of this trial the mean sludge dry solids addition for each liquid type was equal (rates R2 - R4), but the mean metal level applied was significantly different. Thus by analysis of variance, effects due to sludge nutrients (nitrogen and phosphorus) and organic matter could be separated from any metal effects. Maximum mean metal additions achieved in soil in this trial are given in Table 34. Crop yields were not affected according to treatment in 60% of all cases up to these soil metal maxima.

Positive effects on yield occurring mainly in the first season after application accounted for one quarter of all cases (Section 3.1.1). These were probably due to the large quantities of nitrogen added in the sludge, which for example at rate R4 for liquid sludges was approximately 6800 kg N/ha. This was in comparison to recommended levels of

110-210 kg N/ha applied as inorganic fertiliser, to all treatments except rates R4 in 1979 and all treatments in subsequent years to even out manurial sludge effects (Table 5, Section 2.6). Despite this yield increases continued in subsequent years in some cases, and this was probably due to improvements in soil-water holding capacity on the sludged plots, although sensitive crops (lettuce and red beet) were irrigated in dry summers.

Generally there were few effects of reduced yield. Where they did occur, mainly in wheat grain they were probably attributable to excessive sludge nutrients, especially nitrogen. For wheat, however, in contrast to other crops excessive growth led to lodging and subsequent reduction in

Table 34. Maximum levels of phytotoxic elements in soil.

Additions (kg/ha)

	nc		c	
	S1	S4	S1	S4
Ni	42	104	36	116
Cu	162	294	176	380
Zn	353	570	312	666
ZE	1010	1990	952	2350

Estimated increase in concentration (mg/kg)

	Total*				EDTA-extractable+			
	nc		c		nc		c	
	S1	S4	S1	S4	S1	S4	S1	S4
Ni	22	55	24	77	15	34	5	9
Cu	86	157	117	253	63	89	43	68
Zn	188	304	208	444	164	264	96	115
ZE	536	1060	634	1570	418	719	209	329

nc non-calcareous soil; c calcareous loam
S1 liquid sludge types; S4 bed-dried sludge
ZE zinc equivalent (Chumbley 1971)
* conversion factors were nc: 0.56, c: 0.67, see Section 3.2.1
\+ based on figures from Tables 11 and 12.

grain yield. Effects of this type would not be expected in normal operational practice where sludge is added to soil in light dressings in accordance with crop requirements for nutrients.

4.4 Crop composition

Although no yield effects were attributed to sludge metals it was expected that concentrations of metals in crop tissue would demonstrate metal availability from sludged treatments above that seen from unsludged soil. The total Cd level already in the soils before sludging was less than 1 mg/kg, but ranged from 9 - 125 mg/kg for other metals and varied markedly between soil types. The calcareous loam had twice the background Pb content of the non-calcareous soils, half as much Zn and one quarter as much Ni (Table 1, Section 2.2). Metal availability from unsludged treatments is clearly demonstrated by mean metal concentrations in crop tissue sampled from them (Table 35). Thus these values in Table 35 represent some typical background concentrations in crops above which effects of sludge metal availability were judged. Actual concentrations of metals in crops for human consumption may be modified by further processing. For example, boiling vegetables may reduce metal concentrations. Lower concentrations are often observed in samples from dietary surveys where they are prepared as if to be eaten (Davis and Coker 1980).

Critical metal concentrations in certain crops above which yield reductions become significant have been estimated in previous work (Table 36). Also included in the table are the maximum Cd, Ni, Cu, and Zn concentrations attained in crops in this trial. The maxima for this trial did not exceed critical levels in comparative crops except for Cu

Table 35. Background metal concentrations in crops: means for unsludged treatments 1979-83 (mg/kg DM).

Crop	Soil	Cd	Ni	Cu	Zn	Pb
Wheat	sl	0.134	1.5	5.0	29	<=0.8
	c	0.258	1.6	5.9	32	<=0.8
	cl	0.045	<=0.8	5.2	38	<=0.8
Potato	sl	0.148	1.5	5.9	13	<=0.7
	c	0.520	1.2	8.3	15	<=0.7
	cl	0.068	<=0.6	7.2	15	<=0.7
Lettuce	sl	1.00	6.1	11.0	59	1.3
	c	4.67	4.3	18.2	80	2.3
	cl	0.63	1.2	13.1	64	3.9
Red beet	sl	0.286	3.5	7.4	27	<=0.7
	c	0.854	1.6	8.8	31	<=0.7
	cl	0.100	<=1.0	7.6	26	<=1.1
Cabbage	sl	0.114	4.0	3.7	25	<=0.7
	c	0.266	2.0	3.3	23	<=0.7
	cl	0.045	<=0.9	2.8	25	<=1.0
Ryegrass	sl	0.108	3.2	6.4	25	2.0
	c	0.190	3.8	8.4	29	2.4
	cl	0.040	1.7	9.5	35	3.5

Table 36. Upper critical concentrations and maximum values in this trial of phytotoxic metals in crops (mg/kg DM).

Crop	Cd	Ni	Cu	Zn	reference
Upper critical concentrations:					
barley	8	12	19	210	Davis and Beckett (1978)
ryegrass	-	90	22	140	Davis and Carlton-Smith (1984)
Maximum for this trial:					
wheat	1.26 (c)	10 (c)	12 (sl)	120 (c)	
potato	0.89 (c)	4 (sl)	14 (cl)	33 (sl)	
lettuce	15.0 (c)	21 (sl)	35 (c)	355 (c)	
red beet	2.40 (sl)	20 (sl)	23 (c)	150 (c)	
cabbage	1.06 (cl)	15 (c)	8 (c)	94 (cl)	
ryegrass	1.05 (c)	22 (cl)	21 (cl)	100 (sl)	

(sl) sandy loam, (c) clay, (cl) calcareous loam

outlier values due to sample contamination or unmarketable crops excluded.

and Zn in lettuce on clay soil. Thus providing further evidence that cases of yield reduction, which occurred primarily in wheat, were not due to excessive metal uptake. In addition no visible symptoms of metal toxicity were seen in any crop during this trial.

The relationship between crop metal concentration and soil metal was examined on unsludged and liquid sludged treatments for Cd and Zn. Models were adopted which accounted for the maximum average variance in the data thoughout the trial. The model chosen to estimate Cd concentration in crop tissue across a range of Cd concentrations in soil produced by liquid sludge treatment was

$$CT = a + (b \times ST) \tag{10}$$

where CT is the estimated total Cd concentration in crop tissue mg/kg DM

ST is total Cd concentration in the soil mg/kg air-dried soil

a and b, regression coefficient estimates

Parameters a and b for this model could be estimated for all crops on all soils except for Cd in potato on the clay where the relation was not significant. The regression coefficients are summarised in Table 29, Section 3.4.2.

For prediction of Zn in crop tissue the model most suited was

$$CT = a + (b \times \log10(SE)) \tag{11}$$

where SE is the EDTA-extractable Zn concentration in the soil mg/kg air-dried soil

Estimates of a and b were significant in all cases and have been summarised in Table 37.

Table 37. Regression coefficients for equations describing the relation between Zn in crops and EDTA-Zn in soil, unsludged and liquid sludge treated soils.

Crop		sandy loam	clay	calcareous loam
wheat	b	39	33	33
	a	-9	-8	-3
potato	b	6.6	6.7	10
	a	5	6	1
lettuce	b	56	100	38
	a	7	-47	12
redbeet	b	43	55	18
	a	-18	-29	3
cabbage	b	20	23	25
	a	3	-1	-4
ryegrass	b	45	38	31
	a	-16	-5	6

Models were not fitted to Ni and Cu data to relate soil levels to those in the crops. Broad conclusions can be drawn, however, from scatter plots given in earlier reports for this trial and the correlations in Appendix J. Correlations indicated that Ni availability could be modelled in 63 out of the 90 cases of data for all crops. There were significant relationships in at least one season for every crop on every soil, although evidence was very thin for Ni in wheat and red beet on the calcareous loam. Where strong relationships did exist, Ni concentrations in the crop tissue were directly proportional to total Ni in the soil.

Correlations for Cu crop-soil relations indicated that approximately 70% of all cases would justify modelling, the same proportion as with Ni. This was surprising as increases in crop Cu concentration above background levels were relatively small compared to increases seen for other metals. Raw data illustrated in earlier reports indicated many cases where a logarithmic curve would fit best, similar to the model for Zn data.

4.5 Effects of time

In experiments which monitor sewage sludge applied to land by soil analysis, it has been found by many workers that a significant proportion of the metal applied in sludge cannot be accounted for in the sludge-soil profile. Initially in this experiment soil total metal concentrations were higher than expected and subsequently have reduced over four to five years to predicted levels. A gradual mixing of sludge down the soil profile was probably occurring. One clear exception was application of the bed-dried sludge particularly to the clay soil. Here soil analysis showed lower concentrations than expected one year after application and concentrations continued to drop. The sludge appeared to have initially mixed below the sampled depth of 15 cm and continued to move deeper, indicated by reducing levels of all metals by more than ten percent. Alternatively, there may have been lateral movement of sludge-treated soil. There was also evidence of mobility of just Cu and Zn beyond 15 cm from liquid sludge treatments on the calcareous loam.

The second effect of time seen from soil analysis came from EDTA- extractatable metals in soil. The percent of total Cu extracted by EDTA was seen to vary significantly between years on all soils (Section 3.2.2.1). The trend was to increase with time. In addition similar trends were seen for Cu, Ni, and Pb on the two non-calcareous soils. It has already been shown that the proportion of metal extracted by EDTA was far in excess of the crop available portion of any metal (Section 4.2). This change in percent EDTA values with time might, however, be reflecting a change in metal availability to the crops.

Results of Cd availability to wheat and ryegrass in successive years were illustrated in Figures 23 and 24 (Section 3.4.3). Significant seasonal variation did occur in Cd and Zn concentrations in many other crops. The regression coefficient (b) estimated for crop-soil relations in each year for Cd and Zn (Section 4.4), provided a measure of liquid sludge metal which was available to crops. Therefore, in an attempt to identify significant trends with time the b values were correlated with time (year estimated). Correlation coefficients are given for Cd and Zn in Table 38. There were only three significant correlations out of 36: a decrease in Cd availablility to ryegrass on the clay, a decrease in Zn availability to potato on the sandy loam, and an increase in Cd availability to potato on the calcareous loam. These few significant correlations were scattered amongst the set and do not appear to be linked. The only pattern in Table 38 is shown by consistently high positive correlations for both metals on all soils for wheat. None of these values were significant at 95% confidence, however, three were at 90% (r greater than 0.805). In order to substantiate the few trends indicated over the five years further data extending the time period would be necessary.

Table 38 Correlation coefficients to identify trends in liquid sludge metal availability to crops 1979-83.

Crop	Cadmium sl	Cadmium c	Cadmium cl	Zinc sl	Zinc c	Zinc cl
wheat	0.844	0.783	0.763	0.766	0.835	0.820
potato	0.099	-	0.904*	-0.988*	-0.391	-0.390
lettuce	0.151	0.555+	0.700	-0.292	-0.582	-0.102
red beet	-0.661	-0.627	0.253	-0.507	-0.300	0.261
cabbage	0.722+	0.106	0.421	0.139	-0.167	-0.689
ryegrass	-0.512	-0.889*	-0.055	-0.246	-0.703	-0.555

* significant r values ($P < 0.05$)
\+ n = 4, all others n = 5

4.6

Differences between soils

The experiment was carried out on three contrasting soil types in anticipation of significant sludge effects between them. In particular sludge metals studied in the non-calcareous sandy loam and clay soils of average pH 6.6 were expected to be in more soluble forms. They might therefore be more available to crops than in the calcareous loam of pH 8. Other soil characteristics might explain further differences between the non-calcareous soils.

The percentage of EDTA-extractable Ni from sludged plots on the calcareous loam was approximately one fifth of that for the non-calcareous soils. Similarly, half as much Cu and four-tenths as much Zn was extractable by EDTA from the calcareous loam. These relative differences in percent extractable EDTA values between soils might be expected to be reflected in the crop metal concentrations. This was not generally so for Ni or Cu in any crops. Some agreement in relative differences was seen for Zn in red beet but for wheat and ryegrass there was little difference between soils (Figure 22). Mean concentrations of Zn in other crops indicated significantly higher concentrations in lettuce grown on the non-calcareous soils, but little difference for potato and cabbage. Clear differences were indicated only for Zn in lettuce and red beet (bed-dried sludge treatments were excluded from these comparisons).

Cd availability to crops illustrated earlier in Figure 21 showed no significant difference in uptake between the non-calcareous soils by wheat, ryegrass and red beet. Uptake on the calcareous loam, however, was significantly lower for all these three crops. The increase in crop Cd per 1 mg/kg increase in soil Cd due to liquid sludge addition, as estimated by the b coefficient, averaged 0.091 mg/kg in wheat and ryegrass on non-calcareous soils as compared with 0.043 mg/kg on the calcareous loam. Similarly for red beet the crop increases were 0.235 mg/kg and 0.072 mg/kg for non-calcareous and calcareous soils respectively. Cd was also less available from the calcareous loam than the sandy loam to potato. The relation between Cd in potato and Cd in the clay soil was not significant. In lettuce and cabbage significant differences also existed between the non-calcareous soils, Cd was on average 1.6 times more available from liquid sludge, based on b coefficients, on the clay than the sandy loam soil for these crops. Even on the unsludged treatments Cd was more available from clay soil to crops, than from sandy loam (Section 4.4). Soil analysis did not indicate why Cd should be more available on the clay than the sandy loam, reasons for the difference require further investigation. The explanation may lie in the lower content of hydrous oxides of iron and manganese in the clay soil compared with the sandy loam (B J Alloway pers comm). These hydrous oxides contribute to the sorptive capacity of the soil for divalent metal cations.

4.7 Differences between sludges

The two liquid sludges used in the trial were the same type: lagoon-matured digested sludge, and averaged ten percent dry solids. In contrast bed-dried sludge applied to plots was physically quite different with an average of 84% dry solids and a greater proportion of large hard particles. There remained a greater density of visible bed-dried sludge particles, observed at the soil surface after the final cropping season, than liquid sludge particles (Table 39).

EDTA-extractable soil analysis higlighted differences in the extractable proportion of metal between liquid and bed-dried sludge treatments (Table 40).

Table 39. Assessment of visible sludge residues 1984

Sludge type	rate	sandy loam	Soil clay	calcareous loam
none	0	0	0	0
liquid	1	0	0	0
	2	0	0	0
	3	1:5000	1:5000	1:10000
	4	1:1000	1:1000	1:2500
bed-dried	4	1:100	1:200	1:1250
	5	1:200	1:100	1:550

results expressed as estimated proportion of the soil surface occupied by sludge particles.

Table 40. Proportion of total metal extracted by EDTA due to sludge type (percent)

Metal	Liquid sludges		Bed-dried sludge	
	nc	c	nc	c
Ni	68	22	62	12
Cu	74	37	57	27
Zn	87	46	87	26
ZE	78	33	68	21

nc non-calcareous soils
c calcareous loam

Thus on non-calcareous soils %EDTA-Zn values were equal between sludge type, %EDTA-Ni just higher and %EDTA-Cu significantly higher for the liquid sludges. The overall measure for these elements, the Zn equivalent expression, was ten percent higher for liquid sludges. On the calcareous loam greater differences are seen between sludge type for all metals and overall %EDTA-ZE for liquid sludge was twelve percent higher than for bed-dried sludge.

The differences in the EDTA extractable proportion of metal between sludge type could be reflecting differences in crop available fractions. The crop available fraction (CAF) was estimated as follows:

$$CAF = (CTs - CTo)/(STs - STo) \qquad (12)$$

where CTs is the mean metal level in the crop, sludged soil (mg/kg)
CTo the mean metal level in the crop, unsludged soil
STs the mean total metal level in the sludged soil
STo the mean total metal level in the unsludged soil

Values of CAF have been calculated for liquid and bed-dried sludge for Zn (Table 41). Liquid sludge treatments had higher CAF values than bed-dried treatments and this general pattern held true for all soils. This higher crop availability of Zn was also shown by %EDTA values, but only for the calcareous loam. This distinction in CAF values between sludges also varies according to crop which cannot be reflected by EDTA-extractable soil metals.

Table 41. Crop available fraction (CAF) of Zn added to soil according to sludge type.

Crop	Soil	Liquid sludges	(mean)	Bed-dried sludge	(mean)	Mean ratio
wheat	sl	0.39		0.17		
	c	0.20		0.11		
	cl	0.15	(0.25)	0.072	(0.12)	2.1
potato	sl	0.043		0.016		
	c	0.029		0.009		
	cl	0.027	(0.033)	0.009	(0.011)	3.0
lettuce	sl	0.57		0.17		
	c	0.53		0.12		
	cl	0.13	(0.41)	0.05	(0.11)	3.7
red beet	sl	0.40		0.085		
	c	0.40		0.098		
	cl	0.078	(0.29)	0.019	(0.067)	4.3
cabbage	sl	0.15		0.069		
	c	0.17		0.070		
	cl	0.12	(0.15)	0.072	(0.070)	2.1
ryegrass	sl	0.56		0.14		
	c	0.39		0.12		
	cl	0.25	(0.40)	0.10	(0.12)	3.3

Differences also existed in crop availability of Cd between sludges. The crop available fraction of Cd added to soil in liquid sludge was not calculated as for Zn (Equation 12), but determined more accurately by regression analysis. The estimates together with CAF values for bed-dried sludge are given in Table 42. As with Zn, Cd from bed-dried sludge was less available to all crops than Cd from liquid sludges. Differences once more varied between crops: Cd availability from bed-dried sludge was indicated to be a half to an eighth of that from liquid sludges.

Table 42. Crop available fraction of Cd added to soil according to sludge type.

Crop	Soil	Liquid sludge b* estimates	(mean)	Bed-dried sludge CAF+ estimates	(mean)	Mean ratio
wheat	sl	0.097		0.033		
	c	0.105		0.042		
	cl	0.046	(0.083)	0.021	(0.032)	2.6
potato	sl	0.042		0.008		
	c	NS		-		
	cl	0.028	(0.035)	0.007	(0.008)	4.4
lettuce	sl	0.75		0.21		
	c	1.2		0.16		
	cl	0.34	(0.76)	0.13	(0.17)	4.5
red beet	sl	0.22		0.07		
	c	0.24		0.06		
	cl	0.07	(0.18)	0.04	(0.06)	3.0
cabbage	sl	0.034		0.020		
	c	0.051		0.001		
	cl	0.044	(0.043)	0.034	(0.018)	2.4
ryegrass	sl	0.084		0.020		
	c	0.079		0.032		
	cl	0.040	(0.068)	0.026	(0.026)	2.6

* b estimates from regression analyses (Table 29)
\+ CAF crop available fraction calculated from means, Equation 12.

One reason for lower crop availability of Cd and Zn from bed-dried sludge was probably due to the existence of many relatively large sludge particles reducing the likelihood of contact with crop roots.

EDTA-extractable Zn soil analysis which reduces particle size to less than 2 mm showed less difference between bed-dried and liquid sludge than crop measures. Thus metals may also be in less available chemical forms in bed-dried sludge. Physical effects would be expected to decrease with time particularly on cultivated land.

4.8 Soil metal limits

4.8.1 Cadmium

Cadmium applied to agricultural land in sewage sludge has been shown to be taken up by crops. Subsequent transfer of Cd via crops into the food chain could lead to increases in human dietary intake of Cd. The risk to health of Cd and other chemicals in sewage sludge applied to land was recently reviewed by a WHO working group (Dean and Suess 1985). It was concluded that Cd appeared to be the most important contaminant from sewage sludge because of uptake by food plants, and that current limits for Cd in soils and sludge vary greatly between different countries.

Cadmium increases in crops in this trial can be expressed in relation to the current UK guideline limit for Cd in sewage sludge applied to agricultural land (DoE/NWC 1981). The permissible maximum addition of Cd to soil over 30 years is 5 kg/ha. If it is assumed such an addition is incorporated to a depth of 20 cm and soil density equals 1 g/cm^3 then it would lead to an increase of Cd in soil of 2.5 mg/kg (Equation 3 Section 3.2.1) In addition this concentration falls within the range of limit values recommended by the European Communities for Cd (EC 1986). Using b estimates from crop-soil relations (Table 29, Section 3.4.2), increases in crop tissue have been

Table 43. Cadmium increases in crops due to a Cd soil increase of 2.5 mg/kg* from liquid sludge additions.

Crop	% DM	Cd increase in crop tissue				Enrichment factor+	
		mg/kg dry		mg/kg fresh			
		nc	c	nc	c	nc	c
wheat	86	0.25ǂ	0.12	0.22ǂ	0.10	1.3ǂ	2.7
potato	22	0.11ǂ	0.070	0.023ǂ	0.015	1.3ǂ	1.0
lettuce	4	2.4	0.85	0.098	0.034	1.2	1.3
redbeet	15	0.58	0.18	0.086	0.027	1.0	1.8
cabbage	7.7	0.11	0.11	0.008	0.008	0.6	2.4
ryegrass	18	0.20	0.10	0.037	0.018	(1.3)	(2.5)
weighted** average for food crops (excluding ryegrass)						1.2	2.0

* Equivalent to UK maximum permissible addition of 5 kg/ha assuming incorporation to a depth of 20 cm and soil density 1 g/cm^3)
\+ EF, see Equation (13)
nc non-calcareous soils, average for sandy loam and clay
c calcareous loam
ǂ sandy loam only, no significant increase on clay
** according to estimated background dietary intakes of Cd (Hutton and Symon 1985)

estimated according to the UK soil limit (Table 43). As the fraction of Cd available between the non-calcareous sandy loam and clay soils for most crops was similar, an average b estimate was used to estimate Cd increase in crops grown on these soils. To quantify the significance of such increases due to sludge, enrichment factors (EF) expressing them as multiples of Cd concentrations in crops grown on unsludged soil were calculated as follows:

$$EF = b \times 2.5/CTo \quad (13)$$

where b is the regression coefficient estimated for the crop-soil relationship

2.5 mg/kg is the potential increase in Cd concentration in soil

CTo is the mean concentration of Cd in the crop on unsludged soil.

The enrichment factors are included in Table 43, 1.6 - 3.7 times the background concentrations are indicated in crops grown on liquid sludged soil whose Cd concentration was 2.5 mg/kg above background level. The EF values were generally higher for the calcareous loam despite less Cd uptake on this soil. This is because the increases were high relative to their background values (CTo). This illustrates the importance of background levels, which are used to qualify Cd increases in food crops. Different CTo values according to soil type were given earlier (Table 35, Section 4.4).

Crops grown in this trial include the main components of the plant part of the human diet with certain assumptions (Table 44), and amounts eaten have been calculated from statistics for food consumption (MAFF 1983a). By using average amounts eaten for each food group Cd concentrations in crops could simply be converted to 'dietary intake' estimates for each group. This approach was outlined by Davis, Stark and Carlton-Smith (1983) using data from the 1981 season at Royston (calcareous loam). Full evaluation of the relationship between crop concentrations of Cd and human dietary intake of the element would need to take account of various factors including:

i) soil textural classes and land use (types of crop grown) in areas receiving sludge,

ii) Marketing, distribution and preparation of crops grown on sludge-treated land,

iii) background concentrations of Cd in different food crops according to soil type,

Table 44. Plant food components of the human diet and average amounts eaten

MAFF food group/s and number	Crop representing group, this trial	Average amount eaten (kg/day, fresh wt)
1) Bread and cereals	wheat grain	0.240
9) Potatoes	potato	0.175
8C) Leafy salads	lettuce	0.0058
10) Root vegetables and others	red beet	0.103
8A) Cabbage and 8B) legumes	cabbage	0.0727

note: fruit is not included

iv) area of land receiving sludge, rates of application and resulting soil concentrations of Cd,

v) dietary habits of consumers including critical groups,

vi) cadmium availability to crops following heavy one-off applications of sludge as in this trial, compared with repeated light dressings as in operational practice.

Information on the bread and cereals group in particular is most important as it has the largest impact on dietry intake of Cd. These issues have been considered in a report commissioned by WRc on dietary risk assessment for Cd (Hutton and Symon 1985), which could form the basis of further study of the implications of the cadmium content of the crops from Cassington and Royston.

Data from this trial has shown that for an individual who obtained all their plant food from crops grown on liquid sludged soil to produce an increase in soil Cd concentration of 2.5 mg/kg, the proportion of dietary intake of Cd due to plant food (without fruit) would be 2.2 - 3.0 times the intake for an individual obtaining all his crops from unsludged soil. This range was based on weighted average EF values (Table 43) for non-calcareous and calcareous soils, weighted according to the background Cd intakes for each food group estimated by Hutton and Symon (1985). More detailed analysis of dietary implications taking account of the factors outlined above would be of obvious value but is outside the scope of this report.

4.8.2 Nickel, copper and zinc

No reductions in crop yield attributable to the potentially phytotoxic metals were indicated by significance tests using liquid sludge treatments (Section 4.3). The mean concentrations of EDTA-extractable Ni, Cu, and Zn in soil achieved on the liquid sludge treatments were relatively low compared to UK guideline maxima (Table 45), but the concentration ranges were somewhat higher.

When the metals are expressed in terms of the zinc equivalent the mean for the non-calcareous soils in this trial was similar to the guideline maxima of 280 mg/l. Maximum values exceeded this reaching 720 mg/kg for the bed-dried sludge and again were not associated with any discernible effects of metal toxicity in terms of either reductions in yield or visible symptoms.

Table 45. Provisional maximum permissible concentrations of phytotoxic elements in soils treated with sewage sludge in UK and mean values this trial

Element 'extractable'	UK guidelines* (mg/l)		Levels this trial, liquid sludge (mg/kg)			
	non-calc	calc	non-calc		calc	
			range	mean	range	mean
Ni	35	70	5-28	11	<1-6	2
Cu	140	280	8-91	42	3-40	18
Zn	280	560	7-219	92	13-99	48
ZE+	280	560	63-625	264	24-227	100

* levels to be reached in arable soils in 30 years or more (DoE/NWC 1981)
+ zinc equivalent (Chumbley 1971)
'extractable' metals extracted by EDTA
calc calcareous soil

In terms of additions of zinc equivalent, upper rates of sludge application to soil exceeded the maximum permitted in UK guidelines by about 2x and 4x for liquid sludge and bed-dried sludge respectively (Table 34, page 85).

In addition maximum increases of total metal in this trial were comparable to limit values recommended by the European Communities for concentrations of heavy metals in soil (Table 46).

Table 46. Mean total concentrations of phytotoxic elements in comparison to EC limits (mg/kg)

	This trial			EC limit values*
	Minimum (background)	Maxima S1	Maxima S4	
Ni	12	68	79	30 to 75
Cu	17	133	237	50 to 140
Zn	69	376	474	150 to 300

S1 liquid sludge treatments,
S4 bed-dried sludge treatments
* (EC 1986)

Nevertheless no phytotoxic effects of metals were evident in terms of reduced yields or toxic tissue concentrations. The only exceptions to this finding concerned concentrations of copper and zinc in lettuce grown on the clay soil which exceeded upper critical concentrations at high rates of sludge application.

4.8.3 Lead

As with Cd, the principal concern about Pb relates to the potential for increase in dietary intake of the element following the use of sludge on land. Lead concentrations in most edible crops in this trial did not significantly rise above the detection limit of 0.5 - 2.0 mg/kg DM. The only exception to this was lettuce where mean concentrations were approximately 5 mg/kg in several years which expressed on a fresh weight basis would equal 0.2 mg/kg. This is still well below the limit for Pb in human foodstuffs in the UK of 1 mg/kg (fresh weight basis). Generally no significant rise in Pb concentrations in crops occurred above background levels, thus sludge applications would not influence Pb in the human diet by the crop uptake route up to a soil concentration of Pb of at least 330 mg/kg, the highest soil concentration of Pb recorded in the trial. This is in comparison to the EC upper limit of 300 mg/kg (EC 1986).

5. CONCLUSIONS

5.1

Effects of sludge on soil concentrations of metals

1. The amount of cadmium applied in liquid sludge to the top 15 cm of three diverse soil types could be entirely accounted for by soil analyses in this profile, approximately five years after sludge application. The increase in soil metal (Mi) above background values was estimated according to soil density, cultivated depth, and metal loading rate as :

$$Mi = 10 \times Ml/(\rho \times s) \text{ mg/kg}$$

where Ml is the metal loading rate (kg/ha)
ρ the soil density (g/cm^3), air-dried
s the depth of cultivation (cm)

Where the weight of sludge applied exceeds ten percent of the weight of soil in the cultivated layer, then the effect of sludge on soil density should be taken into account.

Amounts of other metals, Ni, Cu, Zn and Pb applied in liquid sludge were also accounted for in the cultivated profile with the exception of Cu and Zn in the calcareous loam. Here some downward movement may have occurred beyond 15 cm depth or there may have been lateral movement of sludge-treated soil off the plots following cultivation.

2. The percentages of EDTA-extractable Ni, Cu, Zn and zinc equivalent (ZE) of total in unsludged soils were lower than for metals of sludge origin:

	unsludged soil			
	Ni	Cu	Zn	ZE
non-calcareous	13	39	6	13
calcareous	4	20	19	12

3. The percentages of EDTA-extractable Ni, Cu, Zn and ZE of total metal of sludge origin for the liquid and bed-dried sludges in non-calcareous and calcareous soil were as follows:

	Liquid sludges				Bed-dried sludge			
	Ni	Cu	Zn	ZE	Ni	Cu	Zn	ZE
non-calcareous	68	74	87	78	62	57	87	68
calcareous	22	37	46	33	12	27	26	21

Crop concentrations of the most plant-available metals (Cd, Zn and Ni) generally followed the pattern of percentage EDTA-extractability being highest on non-calcareous soils and liquid sludge treatments.

5.2
Effects of sludge on crop yields

1. Crop yields were not significantly affected by sludge treatments in 60% of all cases studied.

2. Crop yields were significantly increased in 26% of cases of liquid sludge addition and this was attributed to the beneficial effects on soil structure. Maximum dry solids additions to the soil from liquid sludge was 150 tonnes/hectare and 500 tonnes/hectare for bed-dried sludge.

3. Small (6-10% reductions) but statistically significant reductions in wheat grain yield were seen on the clay and calcareous loam soils treated with liquid sludge and the sandy loam and clay soils treated with bed-dried sludge. The results of comparisons between sludge types suggested this was not due to metals and the most likely explanation was lodging of the crop which occurred due to excessive nitrogen in the soil resulting from the single very heavy application of sludge made in the first year of the trial.

5.3
Effects of sludge on crop composition

1. Increases in metal concentrations in soil due to sludge added, produced significant increases in Cd, Ni, Cu and Zn concentrations in the edible portion of most of the six crops grown: wheat, potato, lettuce, red beet, cabbage and ryegrass.

2. Lead was relatively unavailable to crops from the soils. In the vast majority of cases there was no significant increase of Pb in crop tissue in relation to Pb added to the soil in sludge.

3. Increases in Cd concentrations in crops were most frequently directly proportional to the total Cd concentration in the soil. Thus they were estimated by a linear regression model fitted to Cd in crops plotted against 'total' Cd in soil. Increases in crop concentrations of Cd (mg/kg dm) per 1 mg/kg increase in Cd content of liquid sludge-treated soil (air dry) are summarised below (see also Tables 29 and 42):

Crop	Soil*	Estimated increase in crop tissue concentration per 1 mg/kg increase in soil content.
Wheat grain	sl	0.097
	c	0.105
	cl	0.046
Potato tuber	sl	0.042
	c	NS
	cl	0.028
Lettuce	sl	0.75
	c	1.2
	cl	0.34
Red beet root	sl	0.22
	c	0.24
	cl	0.07
Cabbage	sl	0.034
	c	0.051
	cl	0.044
Ryegrass	sl	0.084
	c	0.079
	cl	0.040

* sl = sandy loam, c = clay, cl = calcareous loam
NS = relationship not statistically significant

4. Increases in Zn concentrations in crops were most frequently proportional to the logarithm of the EDTA-extractable Zn concentration in the soil. Zn levels in crops were best estimated by fitting a logarithmic curve to data when plotted against EDTA-extractable Zn in soil

5.4
Effects of time

1. In general terms agreement between actual and predicted concentrations of metals in soil was best in the last two years of the trial. The explanation for this may be that the repeated cultivations gradually led to better mixing of sludge and soil and hence reduced the error associated with soil sampling.

2. There was little evidence that sludge metals were lost from the cultivated horizon except where sludge was originally incorporated below this depth. Apparent losses of less than ten percent of total Cu and Zn applied to the calcareous loam were indicated. Downward or lateral loss of sludge-treated soil from the plots as a result of cultivation could explain this effect.

3. The EDTA-extractable fraction of total Cu, Ni and Pb from the two non-calcareous soils increased with time.

4. Cadmium and Zn availability to crops over 5 years showed only three definite significant seasonal variations but less than ten percent of cases indicated definite trends over 5 years. Overall there was no trend in crop availability on any soil over 5 years. There was no trend of reducing plant availability of metals over the 5 year period.

5.5 Effects of soil conditions and sludge type

1. The EDTA-extractable fraction of total Ni, Cu and Zn was less from the calcareous loam than the two non-calcareous soils. The relative differences were: Ni one fifth, Cu one half, and Zn four tenths.

2. The rate of increases in Cd concentrations in crops grown on non-calcareous soils was on average double the rate on calcareous soil. This was attributed to the difference in pH of 6.5 for non-calcareous soils in contrast to 8.0 for the calcareous soil.

3. Although the non-calcareous soils had the same pH, Cd was 1.6 times more available from the clay than the sandy loam to lettuce and cabbage. Even on unsludged treatments Cd was most available from the clay. The explanation for this may lie in the low content of iron and manganese sesquioxides of the clay soil.

4. Metals expressed as zinc equivalent were about 10% less extractable from soil treated with bed-dried sludge than from soil treated with liquid sludge (Table 40, Page 95).

5. The availability of metals to crops was in general terms lower in soil treated with bed-dried sludge compared with liquid sludge. This effect depended on the crop in question. Cadmium availability to crops from bed-dried sludge was found to be a half to an eighth of that from liquid sludges (Table 42, Page 97). This was probably because physically discrete bed-dried sludge particles remained in the soil during the trial reducing the likelihood of contact with crop roots. However, EDTA analysis indicated that metals may also be in less available chemical forms. Physical effects would be expected to decrease with time particularly on cultivated land.

5.6

Relation to current recommendations for sludge utilisation

1. Nickel, copper and zinc concentrations in soil produced by the upper rates of liquid and bed-dried sludge were close to the maximum permissible levels set out in the EC Directive for the use of sewage sludge in agriculture. In terms of additions of zinc equivalent, upper rates of sludge application to soil exceeded the maximum permitted in UK guidelines by about 2x

and 4x for liquid sludge and bed-dried sludge respectively (Table 34, Page 85). Nevertheless, no phytotoxic effects of metals were evident in terms of reduced yields, toxic tissue concentrations or visible symptoms. The only exception to this finding concerned concentrations of copper and zinc in lettuce grown on the clay soil which exceeded upper critical concentrations at high rates of sludge application.

2. In general terms there was no significant increase in crop concentrations of lead according to sludge treatment. Sludge applications did not influence the lead content of crops by the plant uptake route up to a soil concentration of Pb of at least 330 mg/kg, the highest recorded in the trial. This is the comparison to the EEC upper limit of 300 mg/kg.

3. In general terms there was a linear relationship between concentrations of cadmium in soil and concentrations of cadmium in the edible parts of the crops studied. An increase of 2.5 mg/kg in the soil concentration of cadmium produced the following increases in crop tissue content of cadmium (mg/kg dm):

Crop	Non-calcareous soils	Calcareous soils
Wheat grain	0.25	0.12
Potato tuber	0.11	0.07
Lettuce	2.4*	0.85
Red beet root	0.58	0.18
Cabbage	0.11*	0.11
Ryegrass	0.20	0.10

* see (viii) 3.

4. The percentage increase in crop cadmium content on calcareous soil was usually less than that seen on the two non-calcareous soils. Extractability of the zinc equivalent metals by EDTA from calcareous soils treated with sludge was about 40% of extractability from non-calcareous soils treated with sludge.

REFERENCES

CHUMBLEY C G (1971) Permissible levels of toxic metals in sewage sludge used on agricultural land. ADAS advisory paper, 10, Pinner, Ministry of Agriculture, Fisheries and Food.

CARLTON-SMITH C H and DAVIS R D (1983) An inter-laboratory comparison of metal determinations in sludge-treated soil. Wat Pollut Control **82**, No 4, 544-556.

COKER E G (1966) Journal of Agricultural Science, Cambridge, **67**, 91-97, 99-103, 105-107.

DAVIS R D and BECKETT P H T (1978) Upper critical levels of toxic elements in plants, II. Critical levels of copper in ground barley, wheat, rape, lettuce and ryegrass and of nickel and zinc in young barley and ryegrass. New phytol **80**, 23-32.

DAVIS R D and CARLTON-SMITH C H (1980) Crops as indicators of the significance of contamination of soil by heavy metals. WRc Technical Report TR 140.

DAVIS R D, STARK J H and CARLTON-SMITH C H (1983) Cadmium in sludge-treated soil in relation to potential human dietary intake of cadmium. Pages 137-146 in Environmental Effects of Organic and Inorganic Contaminants in Sewage Sludge, edited by R Davis et al. D Reidel, Dordrecht.

DAVIS R D and CARLTON-SMITH C H (1984) An investigation into the phytotoxicity of zinc, copper and nickel using sewage sludge of controlled metal content. Environmental Pollution (Series B), **8**, 163-185.

DAVIS R D and COKER E G (1980) Cadmium in agriculture, with special reference to the utilisation of sewage sludge on land. Water Research Centre Technical Report TR 139, 19p.

DEAN R B and SUESS M J (1985) The risk to health of chemicals in sewage sludge applied to land, Waste Management and Research **3**, 251-278.

DEPARTMENT OF THE ENVIRONMENT/NATIONAL WATER COUNCIL (1977) Report of the working party on the disposal of sewage sludge to land. STC Report 5.

DEPARTMENT OF THE ENVIRONMENT/NATIONAL WATER COUNCIL (1981) Report of the sub-committee on the disposal of sewage sludge to land. STC Report 20.

ELLIS (1985) Personal communication.

EUROPEAN COMMUNITIES (1986) Council Directive of 12 June 1986 on the protection of the environment, and in particular of soil, when sewage sludge is used in agriculture (86/278/EEC). Official Journal of the European Communities L181, 29, July.

HALL J E, CARLTON-SMITH C H, DAVIS R D and COKER E G (1983) Field investigations into the manurial value of lagoon-matured digested sewage sludge. WRc report LR 510-M.

HALL J E and COKER E G (1981) Some effects of sewage sludge on soil physical conditions and plant growth. WRc report 130-M.

HUTTON M and SYMON C (1985) Towards a risk assessment for cadmium - Quantifying the soil-plant pathway. Unpublished contract report to WRc.

KERSHAW M A, WOOD R and AINSWORTH G (1962) Sludge treatment and disposal at Maple Lodge. Journal proceedings of the Institute of Sewage Purification **61**, 521-529.

MINISTRY OF AGRICULTURE, FISHERIES AND FOOD (1980) Lime and fertiliser recommendations for vegetables and bulbs. MAFF Publications, Alnwick, Northumberland.

MINISTRY OF AGRICULTURE, FISHERIES AND FOOD (1981a) Lime and fertiliser recommendations for arable crops and grassland. MAFF Publications, Alnwick, Northumberland.

MINISTRY OF AGRICULTURE, FISHERIES AND FOOD (1981b) The analysis of agricultural materials, RB 427, HMSO, London.

MINISTRY OF AGRICULTURE, FISHERIES AND FOOD (1983a) Household food consumption and expenditure: 1981, annual report of the National Food Survey Committee, HMSO, London.

MINISTRY OF AGRICULTURE, FISHERIES AND FOOD (1983b) Survey of cadmium in food: first supplementary reports. Twelfth report of the Steering Group on Food Surveillance, HMSO, London.

MINISTRY OF AGRICULTURE, FISHERIES AND FOOD/AGRICULTURAL DEVELOPMENT AND ADVISORY SERVICE (1978) The determination of cadmium in plant material by direct atomic absorption. Closed conference of analytical chemists, Publications committee.

Ibid (1978) The determination of nitric-perchloric acid soluble cadmium in soil by direct atomic absorption.
Ibid (1976) the determination of cadmium, copper and lead extracted with 0.05 m EDTA, diammonium salt, in soil by atomic absorption.

MINISTRY OF HOUSING AND LOCAL GOVERNMENT, WELSH OFFICE (1970) Taken for granted. Report of the working party on sewage disposal.

DoE STEERING GROUP ON EFFECT OF METALS IN SLUDGE ON CROPS: RELEVANT REPORTS

CHUMBLEY C (1981) Cassington lime recommendations. DoE steering group report No MSC 86.

DEPARTMENT OF THE ENVIRONMENT (1978) Objectives and programme for work at Royston (MSC 35).

JOLLIFFE G (1981) Interim report up to 31 December 1980. DoE steering group report No MSC 73.

JOLLIFFE G (1984) Second interim report up to 31 December 1983. DoE steering group report No MSC 115.

WATSON G G (1979) Effects of metals in sludge on crops: supplementary sludging on certain plots. DoE steering group report No MSC 53.

THAMES WATER/WATER RESEARCH CENTRE PROGRESS REPORTS

THAMES WATER (1978a) Effects of metals in digested sludge on crops. Progress Report to the Department of the Environment. (MSC 27A) February 1977-March 1978.
ibid (1978b) (MSC 34) April 1978-September 1978.
ibid (1978c) (MSC 34 Addendum).
ibid (1979a) (OM/SD/T/1/21) (MSC 44) March 1978-March 1979.
ibid (1979b) (MSC 46) April 1979-June 1979 (Supplement to MSC 44).
ibid (1979c) (MSC 51) April 1979-September 1979.
ibid (1980a) (MSC 56) April 1979-March 1980.
ibid (1980b) (O/Sc/76/2.80) (MSC 67) April 1980-September 1980.
ibid (1981a) (O/Sc/76/121/A81) (MSC 71) April 1980-March 1981 (Part B).
ibid (1981b) (MSC 74) April 1980-March 1981 (Part A).
ibid (1981c) (MSC 85) April 1981-September 1981.
ibid (1982a) (MSC 91) April 1981-March 1982 (Part B).
ibid (1982b) (MSC 97) April 1982-September 1982.
ibid (1982c) (MSC 99) April 1981-March 1982 (Part A).
ibid (1983a) (MSC 102) April 1982-March 1983.
ibid (1983b) (MSC 109) April 1983-September 1983.
ibid (1984) (MSC 116) April 1983-March 1984.

WATER RESEARCH CENTRE (1979a) Effects of metals in sludge on crops: Royston. Progress report to the Department of the Environment 730-S (MSC 41) November 1978-March 1979.
ibid (1979b) (MSC 47) April 1979-June 1979 (Supplement to MSC 41).

ibid (1979c) (MSC 48) April 1979-June 1979 (Additional results).
ibid (1979d) 731-S (MSC 52) April 1979-September 1979.
ibid (1980a) 732-S (MSC 57) October 1979-March 1980.
ibid (1980b) 3-M (MSC 66) April 1980-September 1980.
ibid (1981a) 102-M (MSC 72B) April 1980-March 1981 (Part B).
ibid (1981b) 111-M (MSC 72A) April 1980-March 1981 (Part A). Summary of the first two cropping seasons (1979 and 1980).
ibid (1981c) 213-M (MSC 84) April 1981-September 1981.
ibid (1982a) 318-M (MSC 92) April 1981-March 1982 (Part A). Summary of the first three cropping seasons (1979, 1980 and 1981).
ibid (1982b) 311-M (MSC 93) April 1981-March 1982 (Part B).
ibid (1982c) 420-M (MSC 98) April 1982-September 1982.
ibid (1983a) 510-M (MSC 103) April 1982-March 1983.
ibid (1983b) 655-M (MSC 110) April 1983-October 1983 (Part B).

WATER RESEARCH CENTRE/THAMES WATER (1982) 626-M (MSC 111) April 1982-March 1983 (Part A) Summary of results for 1982 from Cassington and Royston.

WATER RESEARCH CENTRE (1984a) 727-M (MSC 117) April 1983-March 1984 (Part B).

WATER RESEARCH CENTRE/THAMES WATER (1984) 837-M (MSC 121) April 1983-March 1984 (Part A) Summary of results for 1983 for Cassington and Royston.

WATER RESEARCH CENTRE (1984b) 827-M (MSC 122) April 1984-October 1984
ibid (1985) 948-M (MSC 129) April 1984-March 1985.

LIST OF FIGURES AMONGST TEXT

KEY TO ALL FIGURES

symbol	Sludge type
×	background soil only
□	S1 Perry Oaks
○	S2 Hogsmill Valley
▷	S3 S1/S2 mixed
+	S4 bed-dried (excl. from regression)

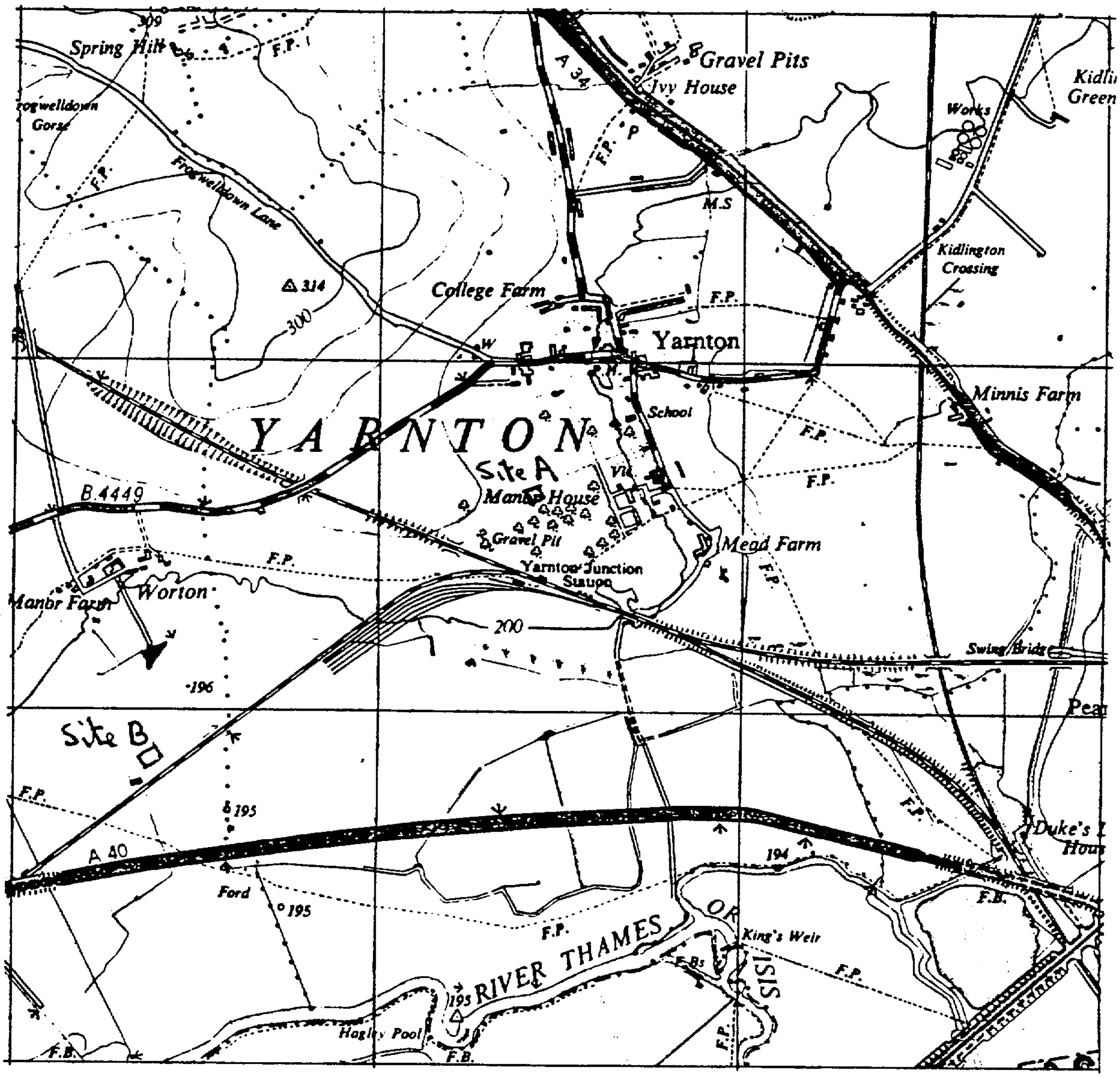
b	slope) or crop mean
a	constant) value : $\bar{y}$
SEb	standard error of slope
r	product-moment correlation coefficient
V%	percent variance accounted for by the regression
n	number of points
- - -	95% confidence limits) for predicted values of Y
CI	± confidence interval)

sl	sandy loam
c	clay
cl	calcareous loam

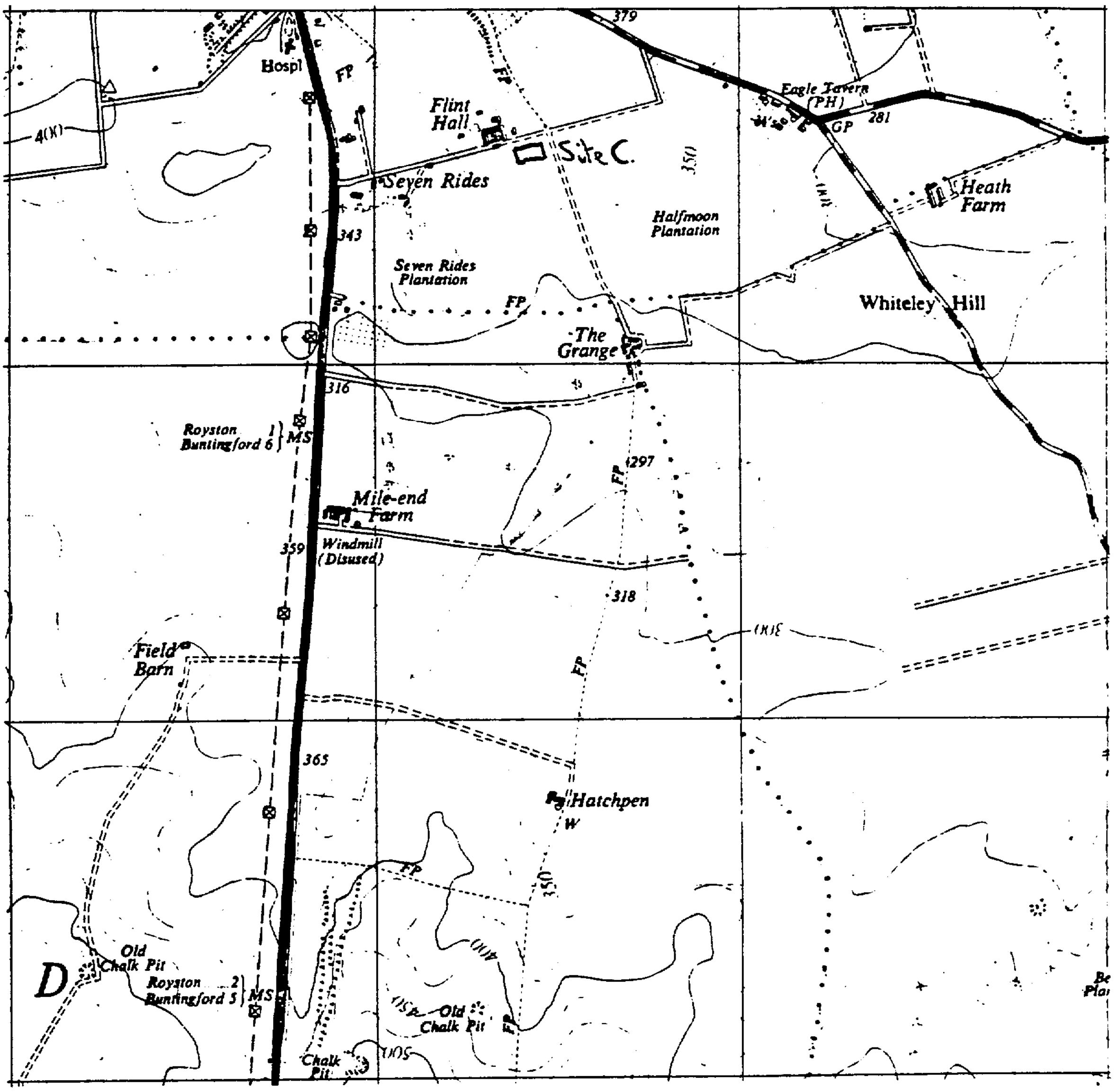
APPENDIX A

SITE LOCATIONS

CASSINGTON SITES A & B (sandy loam and clay)



ROYSTON SITE C (calcareous loam)



APPENDIX B

DETAILS OF FARMING PRACTICES

Crops

In addition to the usual sowing of the five crops occasionally lettuce and cabbage was transplanted from Site A to Site B at Cassington in 1979.

The wheat crop was spring wheat (Timmo 1979) on all three sites and one plot had to be resown in 1980 on Site B. Six plots of ryegrass on Site A were resown in the Spring of 1980. Cabbage and red beet had to be redrilled on Sites A and B in 1982.

Fertilisers

A variety of single and mixed fertilisers were used during the experiment.

Straight potash	as sulphate of potash	
Straight phosphate	super phosphate	
Straight Nitrogen	Nitro chalk 26% N	ICI
	nitrom 33.5% N	ICI
	33.5% N	Norsk Hydro

	N	P	K	
Mixed NPK fertiliser	9	24	24	ICI
	15	15	15	Norsk Hydro
	13	13	20	Fisons' Main Crop
	20	13	20	Fisons' Topgrow

On wheat on all sites Chloromequat (Cycocel) manufactured by Clean Acres was applied to shorten straw.

Chemical sprays

Chemical sprays were used throughout the experiment when necessary.

Crop	Site	Chemical treatment	Trade name	Manufacturer	Target
Rye grass	A	2.4DB	Embutox	May and Baker	Tomato plants
Potatoes	A B	Phorate granules		Chafer	Aphids
	A B	Dinoseb			dessicant for haulms
	A B	Captafol	Sanspor	ICI	Blight
	C	Paraquat, dichloride monolinuron	Gramonol	ICI	Weeds
Winter wheat	A	Mecoprop	CMPP48	Clean Acres	Weeds
	A B	Bromoxynil and ioxynil	Deloxil	Hoechst	Weeds
	A B	Triadimefon	Bayleton	Bayer	Fungicide
	A B C	Cholormequat		Clean Acres	Straw shortener
	C	Dicamba meco propanol	Cambilene	Fisons	Weeds
Cabbage	A B	Pirimiphos-methyl	Sybol 2	ICI	Aphids Caterpillars
	A B	Fonofos and disulphaton			Flea beetles
	C	Oxydemeton-methyl	Metasystox	Bayer	Aphids
	A B	Trifluralin	Treflan	Elanco	Herbicide
		Chlorfenvinphos	Sapecron 10G	Ciba Giegy	Insecticide
		Methiocarb	Draza	Bayer	Slugs
		Heptenophos permethrin	Tumble Bug	Murphy	Insecticide
Red beet	A B	Phorate		Chafer	Insecticide
	A B	Chlorpropham and fenuron	Herbon Yellow	Crop Safe	Herbicide
	A B	Trifluralin	Treflan	Elanco	Herbicide
Lettuce		Metalaxyl	Ridomil	Ciba Giegy	Mildew
		Trifluralin	Treflan	Elanco	
		Propham, diuron, methyl iso propyl phenyl carbamate	Herbon Pink	Crop Safe	Herbicide

APPENDIX C

REGRESSION STATISTICS FOR SOIL ANALYSES

Table C1. a) Correlation coefficients for relation between total metal in soil and metal added in sludge to soil, (liquid sludge treatments).

		Cd	Ni	Cu	Zn	Pb
sandy loam	1979	0.976	0.843	0.959	0.941	0.942
	1980	0.953	0.847	0.930	0.900	(0.903)
	1981	0.978	0.817	0.962	0.939	0.916
	1982	0.962	0.865	0.946	0.931	0.889
	1938	0.958	0.805	0.944	0.905	0.904
clay	1979	0.947	0.896	0.932	0.913	0.928
	1980	0.908	0.759	0.875	0.881	0.879
	1981	0.915	0.841	0.909	0.930	0.914
	1982	0.869	0.767	0.862	0.890	0.743
	1983	0.938	0.902	0.942	0.925	0.937
calcareous loam	1979	0.904	0.832	0.826	0.843	0.535
	1980	0.899	0.900	0.858	0.843	0.722
	1981	0.951	0.917	0.922	0.916	0.629
	1982	0.924	0.911	0.898	0.902	0.644
	1983	0.881	0.830	0.857	0.839	0.545

Table C1. b) Correlation coefficiants for relation between EDTA extractable metal and total metal, (liquid sludge treatments).

		Cd	Ni	Cu	Zn	Pb
sandy loam	1979	0.961	0.845	0.945	0.938	0.922
	1980	0.973	0.857	0.953	0.889	0.949
	1981	0.989	0.764	0.991	0.983	0.975
	1982	0.985	0.895	0.943	0.972	0.974
	1983	0.982	0.784	0.962	0.938	0.960
clay	1979	0.981	0.954	0.974	0.944	0.971
	1980	0.987	0.843	0.956	0.978	0.986
	1981	0.992	0.908	0.979	0.973	0.989
	1982	0.946	0.815	0.962	0.950	0.943
	1983	0.988	0.937	0.989	0.964	0.977
calcareous loam	1979	0.966	0.858	0.928	0.962	0.846
	1980	0.950	0.903	0.930	0.904	0.891
	1981	0.981	0.912	0.963	0.968	0.838
	1982	0.971	0.771	0.949	0.929	0.792
	1983	0.972	0.879	0.960	0.935	0.774

Table C2. Relationship between Ni concentration in soil and Ni added in sludge to soil, liquid sludge treatments.

Soil		1979	1980	1981	1982	1983
sandy loam	slope (b)	0.54	0.64	0.47	0.43	0.44
	SE	0.037	0.033	0.027	0.020	0.026
	constant (a)	36.9	35.4	35.5	32.9	35.7
	SE	0.52	0.50	0.41	0.31	0.40
	n	90	156	156	156	156
clay	b	0.71	0.56	0.68	0.49	0.65
	SE	0.037	0.039	0.035	0.033	0.023
	a	41.9	44.0	42.4	39.6	43.1
	SE	0.53	0.59	0.54	0.51	0.35
	n	90	156	156	156	156
calcareous loam	b	0.91	0.72	0.65	0.63	0.51
	SE	0.041	0.028	0.023	0.023	0.028
	a	11.5	10.9	12.5	11.3	12.8
	SE	0.51	0.37	0.30	0.30	0.37
	n	180	156	155	156	155

units: Ni concentrations in soil mg/kg (Y), Ni added to soil kg/ha (X)
slope: regression coefficient for Y upon X
constant: Y axis intercept
SE: standard error of slope or constant
n: number of points in regression

Table C3. Relationship between Cu concentration in soil and Cu added in sludge to soil, liquid sludge treatments.

Soil		1979	1980	1981	1982	1983
sandy loam	slope (b)	0.56	0.55	0.54	0.45	0.49
	SE	0.018	0.018	0.012	0.013	0.014
	constant (a)	21	23	22.7	22.9	23.5
	SE	1.1	1.2	0.85	0.87	0.95
	n	90	156	156	156	156
clay	b	0.74	0.66	0.75	0.53	0.64
	SE	0.031	0.030	0.028	0.025	0.019
	a	26	27	25	24	27
	SE	2.0	2.0	1.9	1.7	1.3
	n	90	156	156	156	156
calcareous loam	b	0.69	0.61	0.51	0.49	0.45
	SE	0.032	0.030	0.017	0.019	0.022
	a	20	17	20	20	21
	SE	2.0	2.0	1.2	1.3	1.5
	n	180	156	155	156	155

units: Cu concentrations in soil mg/kg (Y), Cu added to soil kg/ha (X)
slope: regression coefficient for Y upon X
constant: Y axis intercept
SE: standard error of slope or constant
n: number of points in regression

Table C4. Relationship between Zn concentration in soil and Zn added in sludge to soil, liquid sludge treatments.

Soil		1979	1980	1981	1982	1983
sandy loam	slope (b)	0.67	0.50	0.53	0.41	0.43
	SE	0.026	0.020	0.016	0.013	0.016
	constant (a)	116	124	121	106	117
	SE	3.8	3.0	2.4	2.0	2.5
	n	90	156	156	156	156
clay	b	0.83	0.65	0.79	0.55	0.64
	SE	0.040	0.028	0.025	0.023	0.021
	a	140	139	135	131	141
	SE	5.7	4.3	3.9	3.5	3.3
	n	90	156	156	156	156
calcareous loam	b	0.80	0.64	0.54	0.51	0.46
	SE	0.035	0.033	0.019	0.020	0.024
	a	64	64	67	67	76
	SE	4.3	4.3	2.5	2.6	3.2
	n	180	156	155	156	155

units: Zn concentrations in soil mg/kg (Y), Zn added to soil kg/ha (X)
slope: regression coefficient for Y upon X
constant: Y axis intercept
SE: standard error of slope or constant
n: number of points in regression

Table C5. Relationship between Pb concentration in soil and Pb added in sludge to soil, liquid sludge treatments.

Soil		1979	1980	1981	1982	1983
sandy loam	slope (b)	0.62	0.57	0.62	0.48	0.53
	SE	0.024	0.022	0.022	0.020	0.020
	constant (a)	29	32	30	30	31
	SE	1.3	1.3	1.3	1.2	1.2
	n	90	156	156	156	156
clay	b	0.80	0.66	0.80	0.50	0.66
	SE	0.034	0.029	0.029	0.037	0.020
	a	35	34	32	32	34
	SE	1.9	1.7	1.7	2.2	1.2
	n	90	156	156	156	156
calcareous loam	b	0.77	0.79	0.60	0.61	0.57
	SE	0.074	0.061	0.060	0.058	0.072
	a	55	53	58	53	53
	SE	2.7	2.3	2.3	2.2	2.8
	n	179	156	155	156	155

units: Pb concentrations in soil mg/kg (Y), Pb added to soil kg/ha (X)
slope: regression coefficient for Y upon X
constant: Y axis intercept
SE: standard error of slope or constant
n: number of points in regression

Table C6. Relationship between EDTA extractable Ni and total Ni in soil, background soil and liquid sludge treatments.

Soil		1979	1980	1981	1982	1983
sandy loam	slope (b)	0.69	0.60	0.79	0.82	0.88
	SE	0.040	0.025	0.041	0.030	0.044
	constant (a)	-21	-17	-23	-22	-25
	SE	1.7	1.1	1.7	1.1	1.8
	n	90	156	156	156	156
clay	b	0.86	0.72	0.72	0.76	0.93
	SE	0.028	0.031	0.025	0.036	0.026
	a	-31	-26	-25	-24	-33
	SE	1.4	1.6	1.2	1.6	1.3
	n	90	156	156	156	156
calcareous loam	b	0.26	0.178	0.150	0.26	0.165
	SE	0.010	0.0062	0.0051	0.014	0.0068
	a	-2.7	-1.7	-1.5	-2.5	-1.4
	SE	0.22	0.12	0.10	0.25	0.12
	n	179	156	155	155	155

Table C7. Relationship between EDTA extractable Zn and total Zn in soil, background soil and liquid sludge treatments.

Soil		1979	1980	1981	1982	1983
sandy loam	slope (b)	0.76	0.88	0.90	0.94	0.99
	SE	0.028	0.032	0.013	0.018	0.028
	constant	-85	-97	-105	-94	-112
	SE	5.7	6.1	2.5	2.9	4.8
	n	90	156	156	156	156
clay	b	0.95	0.91	0.84	0.96	0.95
	SE	0.034	0.015	0.016	0.024	0.020
	a	-123	-117	-109	-119	-124
	SE	8.1	3.5	3.8	4.9	4.5
	n	90	156	156	156	156
calcareous loam	b	0.48	0.39	0.430	0.50	0.44
	SE	0.010	0.013	0.0090	0.017	0.013
	a	-19	-14	-18	-20	-19
	SE	1.6	1.9	1.2	2.0	1.7
	n	178	156	155	156	155

Table C8. Relationship between EDTA extractable Cd and total Cd in soil, background soil and liquid sludge treatments.

Soil		1979	1980	1981	1982	1983
sandy loam	slope (b)	0.82	0.88	0.87	0.90	0.95
	SE	0.24	0.016	0.010	0.013	0.014
	constant (a)	0.0010	0.06	-0.05	-0.01	-0.08
	SE	0.057	0.041	0.025	0.025	0.032
	n	90	156	156	156	156
clay	b	0.84	0.85	0.843	0.87	1.0
	SE	0.018	0.011	0.0088	0.023	0.12
	a	0.12	0.06	0.08	0.07	-0.05
	SE	0.063	0.038	0.032	0.064	0.040
	n	90	156	156	156	156
calcareous loam	b	0.50	0.43	0.483	0.52	0.483
	SE	0.010	0.011	0.0078	0.011	0.0099
	a	-0.01	-0.0003	0.02	-0.08	-0.14
	SE	0.035	0.036	0.022	0.030	0.027
	n	179	156	155	156	155

Table C9. Relationship between EDTA extractable Pb and total Pb in soil, background soil and liquid sludge treatments.

Soil		1979	1980	1981	1982	1983
sandy loam	slope (b)	0.82	0.87	0.85	0.94	0.95
	SE	0.034	0.022	0.015	0.017	0.021
	constant (a)	-15	-15	-15.5	-16.7	-17
	SE	2.0	1.4	0.97	0.96	1.3
	n	90	156	156	156	156
clay	b	0.81	0.87	0.806	0.96	0.92
	SE	0.021	0.012	0.0097	0.026	0.016
	a	-13	-15.8	-12.5	-14	-16
	SE	1.5	0.81	0.74	1.6	1.1
	n	90	156	156	156	156
calcareous loam	b	0.40	0.29	0.32	0.42	0.32
	SE	0.017	0.012	0.015	0.021	0.017
	a	-6	-2	-4	-4	-2
	SE	1.3	1.0	1.1	1.5	1.2
	n	177	154	153	153	154

APPENDIX D

MEAN TOTAL METAL CONCENTRATIONS IN SOIL

Table D1. Mean concentration of Ni in soil, background soil, liquid and bed-dried sludge treatments and theoretical means (mg/kg).

Soil	Sludge		1979	1980	1981	1982	1983	e
sandy loam	none	mean	38.0	34.8	35.7	33.2	36.8	NA
		n	18	36	36	36	36	
	liquid	mean	44.3	44.8	42.2	38.9	41.6	39.1
		n	54	108	108	108	108	
	bed-dried	mean	NA	69.7	64.7	53.7	58.8	74.6
		n	NA	24	24	24	24	
clay	none	mean	42.9	44.7	42.6	39.5	43.3	NA
		n	18	36	36	36	36	
	liquid	mean	51.7	51.8	52.2	46.8	51.6	49.7
		n	54	108	108	108	108	
	bed-dried	mean	NA	73.6	75.9	63.3	69.0	94.5
		n	NA	24	24	24	24	
calcareous loam	none	mean	11.4	11.3	12.6	11.3	12.6	NA
		n	36	36	36	36	36	
	liquid	mean	22.5	19.6	20.5	19.0	18.9	17.4
		n	108	108	108	108	108	
	bed-dried	mean	NA	75.7	62.3	58.7	57.0	78.6
		n	NA	24	24	24	24	

n number of plots/analyses
NA not applicable
e theoretical concentation based on sludge additions (Tables 4 and 5), background soil metals before sludging (Table 1), incorporation to 15 cm depth and soil density 1 g/cm^3, except sandy loam, 1.5 g/cm^3 (see equation (4)).

Table D2. Mean concentration of Cu in soil, background soil, liquid and bed-dried sludge treatments and theoretical means (mg/kg).

Soil	Sludge		1979	1980	1981	1982	1983	e
sandy loam	none	mean	21.3	19.9	21.7	20.4	23.1	NA
		n	18	36	36	36	36	
	liquid	mean	61.3	63.1	61.2	55.7	58.0	55.2
		n	54	108	108	108	108	
	bed-dried	mean	NA	153	140	99.3	121	131
		n	NA	24	24	24	24	
clay	none	mean	25.6.	24.6	24.9	23.2	26.9	NA
		n	18	36	36	36	36	
	liquid	mean	79.2	74.3	79.0	62.5	73.7	70.6
		n	18	36	36	36	36	
	bed-dried	mean	NA	167	157	121	134	177
		n	NA	24	24	24	24	
calcareous loam	none	mean	16.3	15.6	17.0	16.9	17.1	NA
		n	36	36	36	36	36	
	liquid	mean	67.6	56.5	53.8	53.3	50.3	63.2
		n	108	108	108	108	108	
	bed-dried	mean	NA	230	181	171	180	246
		n	NA	24	24	24	24	

n number of plots/analyses
NA not applicable

Table D3. Mean concentration of Zn in soil, background soil, liquid and bed-dried sludge treatments and theoretical means (mg/kg).

Soil	Sludge		1979	1980	1981	1982	1983	e
sandy loam	none	mean	123	116	120	103	119	NA
		n	18	36	36	36	36	
	liquid	mean	223	204	205	172	184	177
		n	54	108	108	108	108	
	bed-dried	mean	NA	357	330	240	284	333
		n	NA	24	24	24	24	
clay	none	mean	143	136	139	131	140	NA
		n	18	36	36	36	36	
	liquid	mean	270	243	260	218	243	230
		n	54	108	108	108	108	
	bed-dried	mean	NA	384	382	309	328	424
		n	NA	24	24	24	24	
calcareous loam	none	mean	69	67	67	68	74	NA
		n	36	36	36	36	36	
	liquid	mean	170	148	139	136	137	147
		n	108	108	108	108	108	
	bed-dried	mean	NA	464	362	359	367	456
		n	NA	24	24	24	24	

n number of plots/analyses
NA not applicable

Table D4. Mean concentration of Pb in soil, background soil, liquid and bed-dried sludge treatments and theoretical means (mg/kg).

Soil	Sludge		1979	1980	1981	1982	1983	e
sandy loam	none	mean	28.7	28.2	27.9	26.8	30.2	NA
		n	18	36	36	36	36	
	liquid	mean	68.4	68.3	69.5	61.3	64.5	52.0
		n	54	108	108	108	108	
	bed-dried	mean	NA	117	125	91.3	104	109
		n	NA	24	24	24	24	
clay	none	mean	32.7	31.3	32.1	28.0	33.5	NA
		n	18	36	36	36	36	
	liquid	mean	85.6	76.5	82.9	64.9	76.2	68.1
		n	54	108	108	108	108	
	bed-dried	mean	NA	125	125	91.9	109	138
		n	NA	24	24	24	24	
calcareous loam	none	mean	57.0	55.8	59.1	53.3	52.8	NA
		n	36	36	36	36	36	
	liquid	mean	85.7	83.7	79.9	75.3	74.3	72.3
		n	108	108	108	108	108	
	bed-dried	mean	NA	192	158	154	158	171
		n	NA	24	24	24	24	

n number of plots/analyses
NA not applicable

APPENDIX E

LIST OF ADDITIONAL DATA AVAILABLE

Table E1. Soil samples, sandy loam.

	Extractable metals: Acetic acid Cd	Ni	Cu	Zn	Pb	Acid NH4 oxalate Cd	Ni	Cu	Zn	Pb	Ammonium acetate Cd	Ni	Cu	Zn	Pb	pH	Other totals Mo	Co	Mn
1979 BLOCK																			
A																			
B																			
C																			
D	*	*	*	*												*			
E	*	*	*	*												*			
F	*	*	*	*	*	–	–	–	–	–	–	–	–	–	–	–	*	*	*
1980																			
A																*			
B																*			
C																*			
D																*			
E																*			
F																*			
1981																			
A																*			
B																*			
C																*			
D																*			
E																*			
F																*			
1982																			
A																*			
B																*			
C																*			
D																*			
E																*			
F																*			
1983																			
A	*	*	*	*	*											*			
B																*			
C																*			
D																*			
E																*			
F	*	*	*	*	*	*	*	*	*	*	*	*	*	*	*	*			

* Data available, usually for all 30 treatments
– – – for cross referencing only.

Table E2. Soil samples, clay.

	Extractable metals																		
	Acetic acid					Acid NH4 oxalate					Ammonium acetate					pH	Other totals		
	Cd	Ni	Cu	Zn	Pb	Cd	Ni	Cu	Zn	Pb	Cd	Ni	Cu	Zn	Pb		Mo	Co	Ag
1979																			
BLOCK																			
A																			
B																			
C																			
D	*	*	*	*												*			
E	*	*	*	*												*			
F	*	*	*	*	-	-	-	-	-	-	-	-	-	-	-	-	*	*	*
1980																			
A																*			
B																*			
C																*			
D																*			
E																*			
F																*			
1981																			
A																*			
B																*			
C																*			
D																*			
E																*			
F																*			
1982																			
A																*			
B																*			
C																*			
D																*			
E																*			
F																*			
1983																			
A	*	*	*	*	*											*			
B																*			
C																*			
D																*			
E																*			
F	*	*	*	*	*	*	*	*	*	*	*	*	*	*	*	*			

Table E3. Soil samples, calcareous loam.

	Extractable metals													
	Acetic acid					Acid NH4 oxalate						pH	CEC	OM%
	Cd	Ni	Cu	Zn	Pb	Cd	Ni	Cu	Zn	Pb	Mo			
1979														
BLOCK														
A	*	*	*	*	*									
B	*	*	*	*	*									
C	*	*	*	*	*									
D	*	*	*	*	*									
E	*	*	*	*	*									
F	*	*	*	*	*									
1980														
A	*	*	*	*	*	*	*	*	*	*	*	*		
B	*	*	*	*	*									
C	*	*	*	*	*	-	-	-	-	-	-	*	*	
D	*	*	*	*	*	-	-	-	-	-	-	*	*	
E	*	*	*	*	*									
F	*	*	*	*	*									
1981														
A	*	*	*	*	*	*	*	*	*	*	*			
B														
C	-	-	-	-	-	-	-	-	-	-	-	-	*	*
D	-	-	-	-	-	-	-	-	-	-	-	-	*	*
E														
F														
1982														
A	*	*	*	*	*	*	*	*	*	*	*			
B														
C	-	-	-	-	-	-	-	-	-	-	-	*	*	*
D	-	-	-	-	-	-	-	-	-	-	-	*	*	*
E														
F														
1983														
A	*	*	*	*	*	*	*	*	*	*	-			
B														
C	-	-	-	-	-	-	-	-	-	-	-	*	*	*
D	-	-	-	-	-	-	-	-	-	-	-	*	*	*
E														
F														

Table E4. Soil samples, calcareous loam (cont'd).

	Extractable metals														
	DTPA					Ammonium nitrate					Sodium nitrate				
	Cd	Ni	Cu	Zn	Pb	Cd	Ni	Cu	Zn	Pb	Cd	Ni	Cu	Zn	Pb
1979															
BLOCK															
A															
B															
C															
D															
E															
1980															
A	*	*	*	*	*	*	*	*	*	*					
B															
C															
D															
E															
1981															
A	*	*	*	*	*										
B															
C															
D															
E															
1982															
A	*	*	*	*	*	–	–	–	–	–	*	*	*	*	*
B															
C															
D															
E															
1983															
A	*	*	*	*	*										
B															
C															
D															
E															

Table E5. Crop samples, sandy loam.

							Other totals					
	Cd	Ni	Cu	Zn	Pb	Mo	Fe	Mn	%Ca	Mg	Ag	As
1979												
BLOCK CROP												
A ryegrass (cut 1)						*	*	*				
ryegrass (cut 2)						*			*	*		
1982												
A ryegrass (cut 1)	-	-	-	-	-	*	-	-	-	-	*	*
ryegrass (cut 2)						*						
ryegrass (cut 3)						*						
ryegrass (cut 4)						*						
D red beet:												
leaves	*	*	*	*	*							
peelings	*	*	*	*	*							
E potato peelings	*	*	*	*	*							
F wheat straw	*	*	*	*	*							
1983												
A ryegrass (cut 1)	-	-	-	-	-	*						
ryegrass (cut 2)	-	-	-	-	-	*						
ryegrass (cut 3)	-	-	-	-	-	*						
ryegrass (cut 4)												
E red beet leaves	*	*	*	*	*							
F potato leaves	*	*	*	*	*							

Table E6. Crop samples, clay.

			Other totals										
			Cd	Ni	Cu	Zn	Pb	Mo	Fe	Mn	Mg	Ag	As
1979													
BLOCK	CROP												
A	ryegrass (cut 1)								*	*	*		
	ryegrass (cut 2)							*	*	*			
1982													
A	ryegrass (cut 1)							*				*	*
	ryegrass (cut 2)							*					
	ryegrass (cut 3)							*					
	ryegrass (cut 4)							*					
D	red beet:												
	leaves		*	*	*	*	*						
	peelings	+	*	*	*	*	*						
E	potato peelings		*	*	*	*	*						
F	wheat straw	+	*	*	*	*	*						
1983													
A	ryegrass (cut 1) - - - - - -							*					
	ryegrass (cut 2) - - - - - -							*					
	ryegrass (cut 3) - - - - - -							*					
	ryegrass (cut 4)												
B	wheat straw		*	*	*	*	*						
E	red beet leaves		*	*	*	*	*						
F	potato leaves		*	*	*	*	*						

+ High and low rates only, S1R0, S1R3, S1R4, S4R4, S4R5.

Table E7. Crop samples, calcareous loam.

		Other totals							
		Cd	Ni	Cu	Zn	Pb	%N	%P	K
1979									
A	ryegrass (cut 1)						*	*	
	ryegrass (cut 2)						*	*	
B	spring wheat:								
	5-leaf	*	*	*	*	*			
	combine harvested	*	*	*	*	*	*	*	
	straw	*	*	*	*	*	*	*	
C	cabbage						*	*	
D	lettuce						*	*	
E	potato peeled						*	*	
F	red beet:								
	leaves	*	*	*	*	*			
	roots (unpeeled)						*	*	
1980									
A	ryegrass (cut 1)						*	*	*
	ryegrass (cut 2)						*	*	*
	ryegrass (cut 3)						*	*	*
	ryegrass (cut 4)						*	*	*
B	potato:								
	peeled						*	*	*
	peelings	*	*	*	*	*			*
C	lettuce						*	*	*
D	cabbage						*	*	*
E	red beet:								
	leaves	*	*	*	*	*			*
	roots						*	*	*
	peel	*	*	*	*	*			*
F	wheat:								
	5 leaf stage	*	*	*	*	*			
	hand picked								*
	combine harvested	*	*	*	*	*	*	*	*
	straw	*	*	*	*	*	*	*	*
1981									
A	ryegrass (cut 1)						*	*	*
	ryegrass (cut 2)						*	*	*
	ryegrass (cut 3)						*	*	*
	ryegrass (cut 4)						*	*	*
B	red beet:								
	root						*	*	
	leaves	*	*	*	*	*			*
	peelings	*	*	*	*	*			*
C	wheat:								
	hand picked								*
	grain combined	*	*	*	*	*	*	*	*
	straw	*	*	*	*	*	*	*	*
	5-leaf	*	*	*	*	*			*

Table E7. Crop samples, calcareous loam (cont'd)

		Other totals							
		Cd	Ni	Cu	Zn	Pb	%N	%P	K
D	lettuce						*	*	*
E	cabbage						*	*	*
F	potato:								
	tubers						*	*	
	peelings	*	*	*	*	*			*
1982									
C	potato peelings	*	*	*	*	*			
D	wheat:								
	5 -leaf	*	*	*	*	*			
	straw	*	*	*	*	*			
F	red beet:								
	leaves	*	*	*	*	*			
	peelings	*	*	*	*	*			
1983									
C	red beet:								
	leaves	*	*	*	*	*			
	peelings	*	*	*	*	*			
D	potato:								
	haulms	*	*	*	*	*			
	peelings	*	*	*	*	*			
E	wheat straw	*	*	*	*	*			

APPENDIX F

DETAILS OF CHEMICAL ANALYSIS OF SAMPLES

CONTENTS

Table F1. Crop metal results reported as equal to or less than the limit of detection.

Cadmium

soil	crop		year	Total no. out of 30 in block
sandy loam	ryegrass	(1)	1979	9
		(2)		10
		(1)	1980	18
		(2)		7
		(1)	1981	13
		(2)		5
		(3)		4
		(4)		5
		(1)	1982	8
		(2)		5
		(3)		3
		(4)		5
		(1)	1983	9
		(2)		8
		(3)		2
		(4)		2
	potato		1979	11
			1981	1
			1982	2
	wheat		1979	15
			1980	2
			1981	4
			1982	2
			1983	1
	cabbage		1980	7
			1981	14
			1982	7
			1983	2
	red beet		1981	1
clay	ryegrass	(1)	1979	1
		(2)		6
		(1)	1980	21
		(2)		1
		(1)	1981	2
		(1)	1982	5
		(2)		4
		(3)		1
		(1)	1983	6
		(2)		6
	wheat		1979	2
			1980	1

Table F1. Crop metal results reported as equal to or less than the limit of detection (cont'd).

Lead

soil	crop	year	Total no. out of 30 in block
sandy loam	ryegrass (2)	1981	1
	potato	1979	30
		1980	26
		1981	22
		1982	16
		1983	29
	wheat	1979	6
		1980	8
		1981	5
		1982	28
		1983	30
	cabbage	1979	10
		1980	21
		1981	17
		1982	25
		1983	19
	lettuce	1980	2
		1981	13
		1982	11
		1983	9
	red beet	1979	30
		1980	20
		1981	29
		1982	19
		1983	23
clay	potato	1979	30
		1980	26
		1981	20
		1982	25
		1983	30
	wheat	1979	4
		1980	17
		1981	9
		1982	25
	cabbage	1979	30
		1980	24
		1981	14
		1982	27
		1983	23

Table F1. Crop metal results reported as equal to or less than the limit of detection (cont'd).

soil	crop	year	Total no. out of 30 in block
clay	lettuce	1980	6
		1981	1
		1982	1
	red beet	1979	26
		1980	9
		1981	17
		1982	12
		1983	23
Nickel			
sandy loam	wheat	1980	1
		1981	7
		1983	1
	potato	1982	2
	red beet	1982	1
clay	potato	1981	2
		1982	1
	wheat	1980	1
		1981	12
		1982	20
	cabbage	1982	1
	red beet	1981	1
		1982	1
		1983	1

Table F2. Data excluded from statistical analysis; not determined or identified as outliers.

Crop data: sandy loam

determinand(s)	crop	year	treatment(s) S	treatment(s) R
Ni	ryegrass	1983	2	0
Cu	wheat	1982	3	3
Zn	ryegrass	1980	2	4
Cd	ryegrass	1979	2	4
			3	4
Pb	ryegrass	1980	4	5
DY, Ni, Cu, Zn, Cd, Pb	lettuce	1980	1	4
Ni, Cu, Zn Cd, Pb	cabbage	1983	1	0
			1	1
			1	3
			1	4
			2	3
			3	0
			3	2
			3	3
			4	5
			4	5

Crop data: clay

determinand(s)	crop	year	treatment(s) S	treatment(s) R
Zn	cabbage	1979	3	0
Cd	potato	1983	1	4
Ni, Cu, Zn, Cd, Pb	lettuce	1983	3	0

DY: dry yield of crop

Table F2. Data excluded from statistical analysis; not determined or identified as outliers (cont'd).

Crop data: calcareous loam

determinand(s)	crop	year	treatment(s) S	treatment(s) R
DY	cabbage	1979	4	5
	lettuce	1983	1	0
			1	0
			1	2
			3	2
Cu	red beet	1982	1	0
Zn	red beet	1979	1	0
Cd	cabbage	1981	2	4

Soil data (strong acid soluble metals): calcareous loam

determinand(s)	block	year	treatment(s) S	treatment(s) R
Cd	B	1981	1	1
Pb	A	1979	2	0
	A		3	2
	C		2	0
	C	1980	2	2
	C		2	3
	D	1981	1	1
	D		3	4
	A	1982	1	4
	B		3	0
	F		1	4
	A	1983	1	4
	E		1	3

Soil data (EDTA extracted metals): calcareous loam

determinand(s)	block	year	treatment(s) S	treatment(s) R
Cu	E	1979	3	4
Zn	E	1979	2	0
Pb	D	1982	1	4
Ni, Cu, Zn, Cd, Pb	F	1979	1	4
	B	1983	1	0

Analysis of cadmium in crops

Subsequent to the work being completed a number of samples of wheat and potato were analysed for Cd by an alternative procedure in view of the high levels of these metals in control samples and in particular for potato when compared to published data (MAFF 1983b). Five grammes of the plant material were digested in nitric/perchloric acids, and the residue taken up in 6M hydrochloric acid. The solution was transferred quantitatively to a separating funnel, made up to about 100 ml and the pH adjusted to about 2 using ammonia solution and thymo-blue as an indicator. This was then shaken with 10 ml of 5% ammonium pyrrolidine dithiocarbamate (APDC) and 20 ml chloroform to extract the metal as the complex. The chloroform layer, containing the metal complex, was then transferred to a clean conical flask. The extraction using fresh APDC and chloroform was repeated twice more and followed by further chloroform extractions. The chloroform was then boiled off and the sample dissolved in nitric/perchloric acids. The solution was taken to dryness, the residue dissolved in 6M hydrochloric acid, and then made up to 10 ml in a graduated tube. This was based on the MAFF method (1981b).

The samples reanalysed were wheat and potato grown on a sandy loam and calcareous loam soils in 1981. Results for Cd were compared to the concentrations determined by the original method by a paired-sample t test (Tables F3 and F4). For Cd in wheat there was no significant difference between the methods on either soil. Concentrations averaged 0.232 mg/kg and 0.164 mg/kg for the sandy loam and calcareous loam soils respectively. For Cd in potato there was a highly significant difference between the methods. On the sandy loam the original method gave results which average

0.201 mg/kg, results for the new procedure were on average 0.047 mg/kg lower than this. On the calcareous soil, however, the original method gave results which averaged 0.080 and results for the new procedure were on average 0.048 higher than this. Results for Cd are also illustrated (Figure F1. The fitted lines according to analytical method would result in similar Cd estimates in crop tissue at around the guideline soil limit of 3 mg/kg. A statistical test comparing the gradient estimates (b) showed that there were no cases where b estimates were significantly different. Results for the bed-dried sludge were not included in the regression analyses and are 'ringed'. Also one obvious outlier for Cd in wheat on the calcareous loam was omitted from the regression.

The relevant laboratories of TW and WRc took part in an inter-laboratory exercise run by the analytical quality assurance sub-group of the Working Party on the Monitoring of Foodstuffs for Heavy Metals in 1983. Their results were found to be of acceptable accuracy for Cd in foodstuffs, as were only six other laboratories out of a total of 28.

Table F3. Comparison of alternative procedure for Cd in wheat 1981 with original method.

a) sandy loam

IDENTIFIER	MINIMUM	MEAN	MAXIMUM	VALUES
New results	0.0500	0.2267	0.5400	30
Old results	0.1000	0.2367	0.6500	30

Plot no.	treatment S	treatment R	old results	new results	Z (new-old)
121	3	3	0.19	0.24	0.05
122	4	6	0.41	0.47	0.06
123	2	3	0.18	0.13	-0.05
124	1	2	0.23	0.22	-0.01
125	2	4	0.21	0.23	0.02
126	1	3	0.35	0.43	0.08
127	2	2	0.10	0.12	0.02
128	1	1	0.20	0.18	-0.02
129	3	0	0.10	0.17	0.07
130	3	2	0.17	0.19	0.02
131	1	4	0.42	0.42	0.00
132	1	0	0.22	0.26	0.04
133	3	4	0.23	0.28	0.05
134	4	5	0.29	0.26	-0.03
135	2	0	0.10	0.06	-0.04
136	1	1	0.21	0.10	-0.11
137	4	5	0.24	0.26	0.02
138	1	2	0.23	0.28	0.05
139	4	6	0.45	0.35	-0.10
140	2	3	0.29	0.21	-0.08
141	3	4	0.32	0.33	0.01
142	3	0	0.13	0.05	-0.08
143	1	4	0.65	0.54	-0.11
144	1	0	0.10	0.10	0.00
145	2	2	0.18	0.13	-0.05
146	3	3	0.18	0.18	0.00
147	1	3	0.30	0.27	-0.03
148	2	4	0.19	0.19	0.00
149	2	0	0.10	0.06	-0.04
150	3	2	0.13	0.09	-0.04

N=30 Mean Z = -0.010 T -1.03 (DF 29) P>0.1
not significant

Table F3. (cont'd)

b) calcareous loam

IDENTIFIER	MINIMUM	MEAN	MAXIMUM	VALUES
New results	0.0400	0.1680	0.6600	30
Old results	0.0100	0.1600	0.6300	30

Plot no.	treatment S	treatment R	old results	new results	Z (new-old)
61	1	3	0.21	0.23	0.02
62	2	4	0.15	0.17	0.02
63	4	6	0.63	0.66	0.03
64	3	4	0.27	0.56	0.29
65	1	3	0.19	0.24	0.05
66	2	0	0.05	0.05	0.00
67	3	3	0.11	0.16	0.05
68	2	4	0.13	0.14	0.01
69	1	0	0.03	0.06	0.03
70	3	2	0.05	0.07	0.02
71	2	2	0.15	0.09	-0.06
72	3	2	0.19	0.20	0.01
73	1	4	0.31	0.32	0.01
74	3	3	0.09	0.08	-0.01
75	3	0	0.01	0.04	0.03
76	1	1	0.19	0.16	-0.03
77	4	6	0.31	0.21	-0.10
78	3	0	0.03	0.04	0.01
79	2	0	0.05	0.04	-0.01
80	2	3	0.11	0.06	-0.05
81	1	0	0.07	0.05	-0.02
82	2	2	0.03	0.06	0.03
83	1	2	0.25	0.17	-0.08
84	3	4	0.21	0.16	-0.05
85	1	2	0.09	0.12	0.03
86	4	5	0.19	0.20	0.01
87	1	4	0.25	0.27	0.02
88	2	3	0.11	0.09	-0.02
89	1	1	0.09	0.11	0.02
90	4	5	0.25	0.23	-0.02

N=30 Mean Z = 0.008 T 0.68 (DF 29) P>0.1
not significant

Table F4. Comparison of alternative procedure for Cd in potato 1981 with original method

a) sandy loam

IDENTIFIER	MINIMUM	MEAN	MAXIMUM	VALUES
New results	0.0700	0.1543	0.2800	30
Old results	0.1000	0.2013	0.3200	30

Plot no.	treatment S	R	old results	new results	Z (new-old)
91	2	0	0.12	0.11	-0.01
92	2	2	0.17	0.14	-0.03
93	4	5	0.28	0.20	-0.08
94	3	0	0.19	0.13	-0.06
95	1	0	0.22	0.14	-0.08
96	3	3	0.19	0.15	-0.04
97	1	2	0.19	0.21	0.02
98	2	4	0.29	0.14	-0.15
99	4	6	0.25	0.17	-0.08
100	3	4	0.26	0.19	-0.07
101	1	1	0.14	0.15	0.01
102	3	2	0.11	0.12	0.01
103	2	3	0.14	0.12	-0.02
104	1	4	0.20	0.21	0.01
105	1	3	0.21	0.13	-0.08
106	1	1	0.24	0.25	0.01
107	1	4	0.32	0.28	-0.04
108	4	6	0.29	0.19	-0.10
109	3	0	0.19	0.15	-0.04
110	4	5	0.20	0.17	-0.03
111	2	3	0.17	0.13	-0.04
112	3	3	0.24	0.19	-0.05
113	3	2	0.26	0.18	-0.08
114	1	3	0.29	0.23	-0.06
115	1	0	0.30	0.14	-0.16
116	3	4	0.11	0.10	-0.01
117	2	0	0.10	0.08	-0.02
118	2	4	0.11	0.08	-0.03
119	1	2	0.16	0.08	-0.08
120	2	2	0.10	0.07	-0.03

N = 30 Mean Z = -0.047 T -5.83 (DF 29) P<0.001
very highly significant

Table F4. (cont'd)

b) calcareous loam

IDENTIFIER	MINIMUM	MEAN	MAXIMUM	VALUES
New results	0.0500	0.1277	0.2200	30
Old results	0.0000	0.0800	0.2000	30

Plot no.	treatment S	treatment R	old results	new results	Z (new-old)
151	3	4	0.10	0.12	0.02
152	1	0	0.04	0.07	0.03
153	4	5	0.08	0.16	0.08
154	1	2	0.12	0.19	0.07
155	2	3	0.08	0.11	0.03
156	4	6	0.08	0.16	0.08
157	1	1	0.16	0.17	0.01
158	2	0	0.00	0.05	0.05
159	3	0	0.10	0.07	-0.03
160	3	4	0.08	0.13	0.05
161	1	3	0.20	0.19	-0.01
162	3	3	0.06	0.15	0.09
163	2	4	0.02	0.08	0.06
164	1	4	0.16	0.22	0.06
165	2	4	0.08	0.18	0.10
166	1	0	0.04	0.06	0.02
167	2	3	0.02	0.08	0.06
168	1	2	0.08	0.16	0.08
169	1	1	0.12	0.15	0.03
170	4	5	0.12	0.15	0.03
171	3	3	0.06	0.14	0.08
172	1	4	0.12	0.20	0.08
173	3	0	0.04	0.08	0.04
174	2	0	0.00	0.06	0.06
175	2	2	0.04	0.09	0.05
176	4	6	0.12	0.14	0.02
177	1	3	0.10	0.14	0.04
178	2	2	0.04	0.10	0.06
179	3	2	0.06	0.11	0.05
180	3	2	0.08	0.12	0.04

ZBAR 0.048 T 8.80 DF 29

N = 30 Mean Z = 0.048 T 8.80 (DF 29) P<0.001
very highly significant

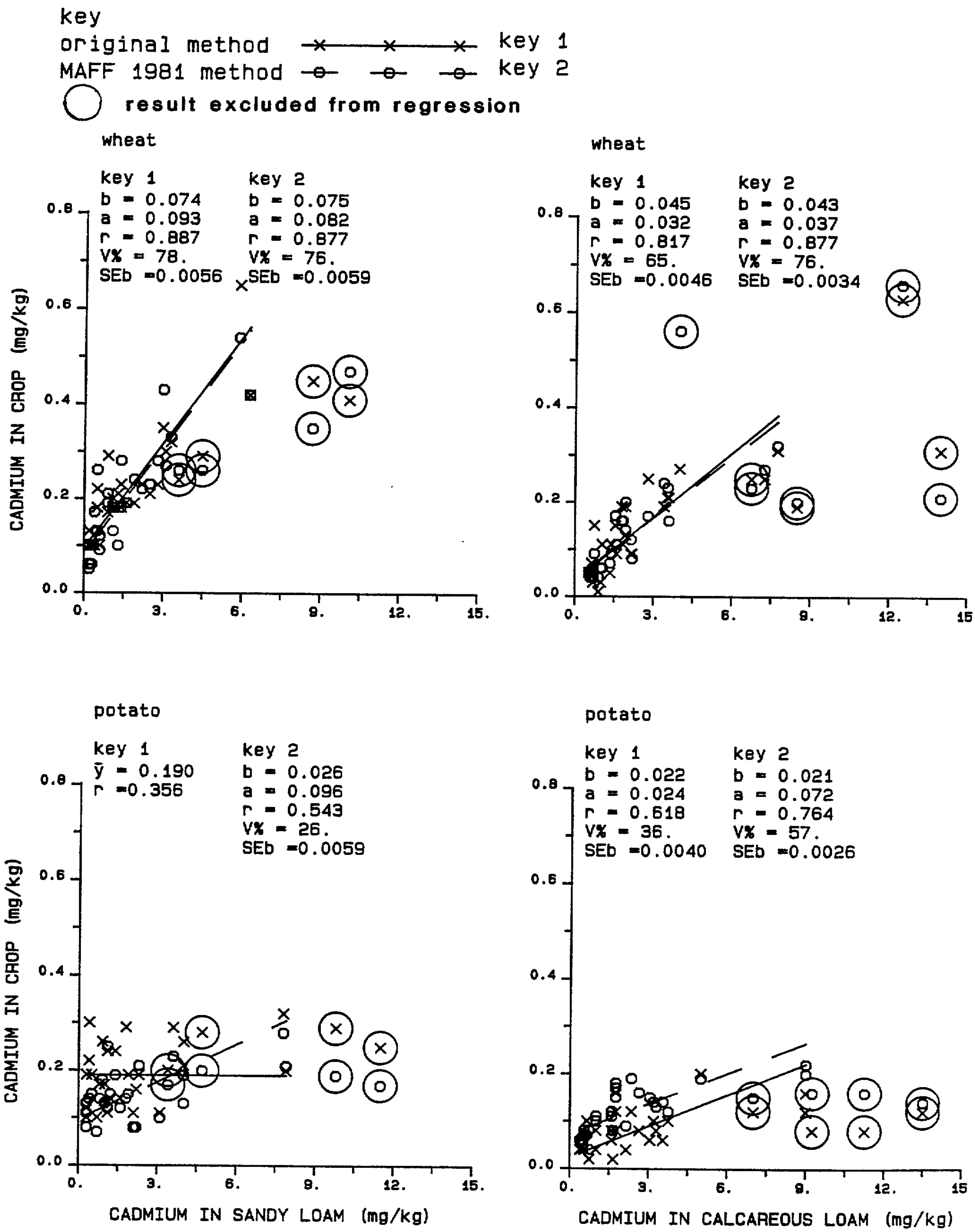


Fig F1. Comparison between analytical methods for the determination of cadmium in crops.

Table F5. Limits of detection for determination of metals in crop and soil samples (mg/kg)

Metal	Crop samples			Soil samples			
				'total'		EDTA	
	nc	c	MAFF (1981c*)	nc	c	nc	c
Cd	0.1	0.1	0.02	0.25	0.15	0.2	0.1
Ni	0.4	1	-	1.2	2	2	1
Cu	0.2	0.4	-	0.5	1	0.2	0.2
Za	0.2	0.2	-	0.4	0.5	0.2	0.5
Pb	0.5	2	0.2	1.2	5	2	2

nc non-calcareous soils, c calcareous loam

Notes: electrodeless discharge lamp used for Cd on calcareous soil

limits of detection were higher when measurable concentrations were present in blanks.

* alternative procedure for analysis of Cd and Pb in plant material used for reanalysis of wheat and potato, see text, this Appendix.

Method for the preparation of plant materials for the determination of Cd, Cu, Ni, Pb and Zn by atomic absorption spectrometry at WRc.

Principle	Plant material is brought into solution and the organic matter destroyed by digestion with concentrated nitric acid.
Apparatus	Boiling tubes, graduated at 25.0 ml, with ground glass stoppers. Thermostatically controlled heating block, with holes drilled to the appropriate size for the boiling tubes. Vortex mixer. Top loading balance, 3 decimal places.
Reagents	Nitric acid d 1.42. Water, distilled or deionised. Anti-bumping granules.
Procedure	Weigh 1.250 $\pm$ 0.002 g sample into a clean, dry boiling tube and add about 3 anti-bumping granules. Carefully add 1 ml of nitric acid down the side of the tube then make further additions until the sample is wetted, using the Vortex mixer if necessary. Leave overnight for the initial reactions to subside. Add further nitric acid until 7 ml in all has been used. Heat the tube gently (about 50 ^{o}C) in the thermostatically controlled heating block, making sure that none of the sample is expelled from the tube by using the mixer and cooling if necessary.

When the reaction subsides, raise the temperature of the block until the solution is boiling steadily, the tube acting as an air condenser, and continue to boil until the undissolved sample is a homogeneous colour. Raise the temperature of the block until most of the nitric acid has been driven off, but a small volume, about 1 ml, remains, taking care not to raise the temperature of the block too high or bumping will result.

Cool the tube, allowing fats, if present, to solidify, then make the solution up to the graduation mark. Stopper the tube, then invert it to make the solution homogeneous. The resulting solution will be clear, with the remaining solids settling out quickly onto the bottom of the tube. Analyse the supernatent solution.

Notes:
a) Blanks should be run with each batch of samples.
b) As the concentration of acid in these solutions is relatively high and variable check solutions of known concentration should be run through the entire procedure.
c) This method can also be used for soils, although it is more appropriate to take a smaller weight of sample. About 0.5 g has been shown to be suitable.

Method for the determination of Cd, Cu, Ni, Pb and Zn in plant materials by atomic absorption spectrometry by WRc.

Principle — Metals with nitric acid are determined by comparison with standards by atomic absorption spectrometry.

Apparatus — Atomic absorption spectrometer.

Reagents — Standard solutions of 1000 µg/ml of Cd, Cu, Ni, Pb and Zn, prepared from 'Suprapure' material. Commercially available standards may be used if they have been shown to have the correct concentration of the element in solution.

Procedure — Prepare standards and check solutions of the following concentrations.

Cd	1.00	µg/ml	in	0.1 m	HNO_3	0.50	µg/ml	in	0.5 m	HWO_3
Cu	4.00	"	"	"	"	2.00	"	"	"	"
Ni	4.00	"	"	"	"	2.00	"	"	"	"
Pb	5.00	"	"	"	"	1.00	"	"	"	"
Zn	1.00	"	"	"	"	0.50	"	"	"	"

Set up the instrument according to the manufacturer's instructions using the following wavelengths. The background corrector must be used for all elements.

Cd 228.8 nm
Cu 324.8 nm
Ni 232.0 nm
Pb 283.3 nm
Zn 213.9 nm

Note on the worksheet the gasflows, burner height, indicated wavelength, lamp current, slit width and absorbance for the calibration standard. Set the appropriate integration time. Run the calibration blank and standard and set to the corresponding values. Nebulise the samples and note the readings. Dilute the samples, if necessary, with 0.5 M HNO_3 to bring them within the calibration range. The check solutions should be used in the course of the determinations to ensure that appropriate accuracy and precision is being achieved.

Calculation Multiply the blank corrected concentration in µg/ml by the volume of sample and divide by the sample weight to give the concentration in µg/g.

$$\text{ie } C = (R-\bar{B})\frac{V}{W}$$

where C = concentration of element in sample in µg/g

R = concentration of element in solution in µg/l

$\bar{B}$ = mean of blanks in µg/l

V = volume of solution in ml

W = weight of sample in grams.

Note: This method can also be used for the determination of metals in soils and sludge - amended soils prepared by the nitric acid procedure.

APPENDIX G -

MEAN YIELDS OF CROPS

Table G1. Mean wheat yields (t/ha DM)

Soil	Sludge		1979	1980	1981	1982	1983	SE
sandy loam	none	mean ($\bar{x}$)	3.69	4.30	6.98	5.76	5.13	0.264
	liquid	$\bar{x}$	4.27	3.91	6.37	5.32	4.98	0.152
	bed-dried	$\bar{x}$	NA	3.88	5.88	5.20	3.39	0.329
clay	none	$\bar{x}$	1.30	4.08	7.30	6.77	5.18	0.227
	liquid	$\bar{x}$	2.10	3.99	6.97	4.92	3.91	0.131
	bed-dried	$\bar{x}$	NA	3.78	7.03	4.53	3.21	0.263
calcareous loam	none	$\bar{x}$	2.72	4.43	5.25	5.54	5.45	0.251
	liquid	$\bar{x}$	2.70	3.83	5.18	5.39	4.88	0.145
	bed-dried	$\bar{x}$	NA	4.04	4.60	5.28	3.64	0.255

Table G2. Mean potato yields (t/ha DM)

Soil	Sludge		1979	1980	1981	1982	1983	SE
sandy loam	none	mean ($\bar{x}$)	9.36	8.58	13.65	9.92	4.53	0.428
	liquid	$\bar{x}$	10.43	8.86	13.34	8.77	3.96	0.247
	bed-dried	$\bar{x}$	NA	5.38	12.83	8.95	3.19	0.449
clay	none	$\bar{x}$	11.90	10.67	8.47	10.06	-	0.562
	liquid	$\bar{x}$	11.51	9.01	10.28	9.35	-	0.324
	bed-dried	$\bar{x}$	NA	7.92	14.05	8.91	-	0.574
calcareous loam	none	$\bar{x}$	7.63	6.71	9.14	7.45	10.50	0.374
	liquid	$\bar{x}$	9.08	8.75	10.39	8.25	10.33	0.216
	bed-dried	$\bar{x}$	NA	7.49	10.78	8.26	10.71	0.568

Table G3. Mean lettuce yields (t/ha DM)

Soil	Sludge		1979	1980	1981	1982	1983	SE
sandy loam	none	mean ($\bar{x}$)	0.82	1.00	1.60	1.65	1.39	0.087
	liquid	$\bar{x}$	1.08	1.01	1.88	1.58	1.37	0.050
	bed-dried	$\bar{x}$	NA	0.83	1.85	1.64	1.32	0.111
clay	none	$\bar{x}$	1.24	1.37	1.45	1.50	-	0.083
	liquid	$\bar{x}$	1.42	1.48	1.72	1.54	-	0.048
	bed-dried	$\bar{x}$	NA	1.40	1.60	1.63	-	0.110
calcareous loam	none	$\bar{x}$	0.33	0.88	0.92	1.66	0.76	0.106
	liquid	$\bar{x}$	0.74	1.33	1.33	1.53	0.91	0.061
	bed-dried	$\bar{x}$	NA	1.02	1.02	1.84	1.01	0.137

Table G4. Mean cabbage yields (t/ha DM)

Soil	Sludge		1979	1980	1981	1982	1983	SE
sandy loam	none	mean ($\bar{x}$)	6.07	8.42	8.10	8.12	5.77	0.383
	liquid	$\bar{x}$	7.23	8.09	9.24	8.02	6.54	0.221
	bed-dried	$\bar{x}$	NA	7.30	8.73	7.92	6.63	0.639
clay	none	$\bar{x}$	7.65	6.99	11.58	8.70	8.98	0.367
	liquid	$\bar{x}$	7.96	5.83	11.74	9.59	10.22	0.212
	bed-dried	$\bar{x}$	NA	6.96	12.73	8.27	9.16	0.526
calcareous loam	none	$\bar{x}$	4.91	5.25	7.70	8.54	5.41	0.392
	liquid	$\bar{x}$	8.23	6.02	8.28	9.45	6.19	0.226
	bed-dried	$\bar{x}$	NA	4.88	6.96	9.76	5.80	0.261

Table G5. Mean red beet yields (t/ha DM)

Soil	Sludge		1979	1980	1981	1982	1983	SE
sandy loam	none	mean ($\bar{x}$)	5.33	6.75	7.30	4.34	5.53	0.360
	liquid	$\bar{x}$	6.83	5.61	7.81	3.94	4.91	0.208
	bed-dried	$\bar{x}$	NA	4.65	9.53	4.40	5.65	0.458
clay	none	$\bar{x}$	5.41	6.20	8.82	6.68	5.92	0.358
	liquid	$\bar{x}$	5.49	5.81	9.02	6.40	5.40.	0.207
	bed-dried	$\bar{x}$	NA	5.85	9.95	6.65	5.74	0.449
calcareous loam	none	$\bar{x}$	3.02	5.43	6.00	8.07	9.17	0.430
	liquid	$\bar{x}$	5.48	6.44	7.37	8.69	10.11	0.248
	bed-dried	$\bar{x}$	NA	4.57	8.85	8.41	11.74	0.656

Table G6. Mean ryegrass yields (t/ha DM)

Soil	Sludge		1979	1980	1981	1982	1983	SE
sandy loam	none	mean($\bar{x}$)	3.00	8.35	11.37	7.12	10.33	0.299
	liquid	$\bar{x}$	5.80	8.90	11.76	8.67	10.06	0.173
	bed-dried	$\bar{x}$	NA	2.85	12.05	9.53	10.00	0.585
clay	none	$\bar{x}$	5.97	10.88	12.67	12.06	11.27	0.494
	liquid	$\bar{x}$	7.21	12.59	13.16	12.14	9.37	0.285
	bed-dried	$\bar{x}$	NA	7.30	14.30	12.61	8.80	0.863
calcareous loam	none	$\bar{x}$	1.01	13.29	14.08	12.67	11.09	0.537
	liquid	$\bar{x}$	3.07	15.76	13.66	11.97	10.24	0.310
	bed-dried	$\bar{x}$	NA	8.55	13.89	13.24	10.03	1.036

APPENDIX H

CORRELATIONS BETWEEN YIELD, SLUDGE APPLICATIONS AND SOIL METALS

Table H1. Correlations between yield and sludge applications, wheat and potato on sandy loam soil.

		Wheat ds	Wheat Ni	Wheat Cu	Wheat Zn	Potato ds	Potato Ni	Potato Cu	Potato Zn
1979		0.090				0.367			
	Add.		0.208	0.155	0.150		0.457	0.432	0.429
	Con.		0.179	0.193	0.176		0.383	0.433	0.460
	EDTA		0.252	0.269	0.236		0.408	0.404	0.419
1980		-0.211				0.016			
	Add.		-0.052	-0.141	-0.145		0.303	0.165	0.156
	Con.		0.154	-0.044	-0.105		0.218	0.078	0.078
	EDTA		0.028	-0.112	-0.085		0.292	0.190	0.104
1981		-0.356				0.068			
	Add.		-0.092	-0.239	-0.246		0.053	0.065	0.065
	Con.		-0.112	-0.315	-0.337		-0.118	0.015	-0.008
	EDTA		-0.124	-0.268	-0.314		0.054	0.011	0.036
1982		-0.700				-0.467			
	Add.		-0.441	-0.600	-0.609		-0.306	-0.409	-0.412
	Con.		-0.418	-0.641	-0.646		-0.347	-0.451	-0.426
	EDTA		-0.482	-0.655	-0.700		-0.339	-0.452	-0.426
1983		-0.589				-0.223			
	Add.		-0.535	-0.589	-0.593		-0.137	-0.191	-0.194
	Con.		-0.227	-0.550	-0.448		-0.034	-0.186	-0.122
	EDTA		-0.509	-0.541	-0.671		-0.129	-0.202	-0.178

Table H2. Correlations between yield and sludge applications, cabbage and lettuce on sandy loam soil.

Year		Cabbage ds	Cabbage Ni	Cabbage Cu	Cabbage Zn	Lettuce ds	Lettuce Ni	Lettuce Cu	Lettuce Zn
1979		0.382				0.591			
	Add.		0.360	0.391	0.390		0.436	0.542	0.545
	Con.		0.001	0.375	0.293		0.404	0.583	0.562
	EDTA		0.351	0.377	0.366		0.405	0.561	0.514
1980		-0.239				0.371			
	Add.		-0.030	-0.143	-0.150		0.191	0.304	0.310
	Con.		-0.102	-0.109	-0.119		0.186	0.237	0.171
	EDTA		-0.049	-0.122	-0.249		0.124	0.234	0.257
1981		0.420				0.234			
	Add.		0.461	0.465	0.461		0.045	0.149	0.155
	Con.		0.506	0.471	0.473		0.027	0.228	0.230
	EDTA		0.388	0.433	0.473		0.137	0.250	0.205
1982		0.224				0.255			
	Add.		0.181	0.211	0.214		0.395	0.343	0.336
	Con.		0.220	0.160	0.195		0.336	0.283	0.277
	EDTA		0.103	0.144	0.133		0.335	0.284	0.300
1983		0.347				0.107			
	Add.		0.219	0.298	0.302		0.118	0.118	0.118
	Con.		1.055	0.886	0.892		0.041	0.195	0.258
	EDTA		0.936	0.895	0.887		0.139	0.057	0.156

Table H3. Correlations between yield and sludge applications, ryegrass and red beet on sandy loam soil.

Year	Ryegrass ds		Ni	Cu	Zn	Red beet ds	Ni	Cu	Zn
1979	0.793					0.635			
		Add.	0.619	0.745	0.749		0.727	0.715	0.712
		Con.	0.484	0.727	0.678		0.781	0.691	0.707
		EDTA	0.562	0.714	0.695		0.712	0.666	0.627
1980	0.340					0.083			
		Add.	0.160	0.264	0.271		0.012	0.051	0.052
		Con.	0.257	0.168	0.152		0.131	0.142	0.202
		EDTA	0.116	0.139	0.048		0.098	0.185	0.116
1981	0.215					0.284			
		Add.	0.359	0.300	0.295		0.251	0.281	0.282
		Con.	0.363	0.232	0.177		0.192	0.246	0.246
		EDTA	0.458	0.231	0.197		0.173	0.233	0.218
1982	0.700					0.152			
		Add.	0.609	0.689	0.692		0.012	0.074	0.079
		Con.	0.587	0.728	0.693		0.094	0.096	0.062
		EDTA	0.621	0.613	0.716		0.013	0.085	0.070
1983	0.487					0.364			
		Add.	0.383	0.458	0.461		0.433	0.417	0.415
		Con.	0.411	0.430	0.477		0.250	0.333	0.343
		EDTA	0.381	0.445	0.461		0.397	0.348	0.337

Table H4. Correlations between yield and sludge applications, wheat and potato on clay soil.

Year	Wheat ds		Ni	Cu	Zn	Potato ds	Ni	Cu	Zn
1979	0.519					0.044			
		Add.	0.378	0.473	0.477		0.248	0.151	0.145
		Con.	0.285	0.465	0.457		0.323	0.214	0.219
		EDTA	0.342	0.502	0.482		0.292	0.183	0.139
1980	-0.224					-0.047			
		Add.	-0.025	-0.132	-0.139		0.106	0.030	0.026
		Con.	0.322	-0.080	-0.125		0.033	0.039	0.069
		EDTA	0.023	-0.086	-0.129		0.130	0.066	0.084
1981	-0.550					0.587			
		Add.	-0.336	-0.466	-0.474		0.530	0.588	0.589
		Con.	-0.272	-0.424	-0.393		0.484	0.616	0.586
		EDTA	-0.303	-0.442	-0.451		0.619	0.556	0.627
1982	-0.848					-0.428			
		Add.	-0.630	-0.779	-0.785		-0.540	-0.508	-0.504
		Con.	-0.758	-0.834	-0.808		-0.388	-0.452	-0.422
		EDTA	-0.678	-0.843	-0.841		-0.484	-0.441	-0.406
1983	-0.850					*			
		Add.	-0.653	-0.792	-0.797		*	*	*
		Con.	-0.439	-0.776	-0.747		*	*	*
		EDTA	-0.643	-0.776	-0.755		*	*	*

Table H5. Correlations between yield and sludge applications, cabbage and lettuce on clay soil.

	Cabbage ds		Ni	Cu	Zn	Lettuce ds	Ni	Cu	Zn
1979	0.142					0.428			
		Add.	0.017	0.085	0.089		0.332	0.401	0.403
		Con.	0.005	0.129	0.141		0.279	0.350	0.431
		EDTA	0.033	0.158	0.158		0.284	0.351	0.375
1980	0.429					0.126			
		Add.	0.221	0.344	0.350		0.106	0.120	0.122
		Con.	0.155	0.306	0.349		0.056	0.093	0.099
		EDTA	0.227	0.317	0.327		0.082	0.037	0.100
1981	0.084					0.402			
		Add.	0.107	0.100	0.100		0.318	0.379	0.380
		Con.	0.363	0.191	0.143		0.224	0.298	0.314
		EDTA	0.197	0.172	0.150		0.266	0.328	0.326
1982	0.617					0.151			
		Add.	0.468	0.572	0.576		0.019	0.071	0.076
		Con.	0.399	0.543	0.527		0.037	0.109	0.147
		EDTA	0.447	0.557	0.576		0.034	0.128	0.162
1983	0.268					*			
		Add.	0.158	0.226	0.229		*	*	*
		Con.	0.099	0.173	0.042		*	*	*
		EDTA	0.130	0.225	0.177		*	*	*

Table H6. Correlation between yield and sludge applications, ryegrass and red beet on clay soil.

Year	Ryegrass ds		Ni	Cu	Zn	Red beet ds	Ni	Cu	Zn
1979	0.306					0.022			
		Add.	0.356	0.348	0.345		0.181	0.106	0.101
		Con.	0.342	0.410	0.380		0.262	0.151	0.145
		EDTA	0.415	0.454	0.423		0.235	0.139	0.128
1980	0.084					0.052			
		Add.	0.115	0.106	0.103		0.006	0.031	0.031
		Con.	0.148	0.193	0.181		0.038	0.029	0.001
		EDTA	0.151	0.177	0.160		0.084	0.054	0.111
1981	-0.311					0.066			
		Add.	-0.075	-0.203	-0.212		0.230	0.084	0.074
		Con.	-0.010	-0.275	-0.304		0.115	0.025	0.073
		EDTA	-0.129	-0.284	-0.323		0.163	0.014	0.064
1982	0.015					0.050			
		Add.	-0.192	-0.091	-0.085		0.180	0.118	0.115
		Con.	-0.387	-0.146	-0.149		0.052	0.059	0.007
		EDTA	-0.199	-0.116	-0.063		0.093	0.086	0.085
1983	-0.813					0.108			
		Add.	-0.669	-0.781	-0.784		0.224	0.175	0.170
		Con.	-0.655	-0.786	-0.812		0.325	0.201	0.179
		EDTA	-0.670	-0.797	-0.836		0.255	0.186	0.173

Table H7. Correlations between yield and sludge appications, wheat and potato on calcareous loam soil.

	Wheat ds		Wheat Ni	Wheat Cu	Wheat Zn	Potato ds	Potato Ni	Potato Cu	Potato Zn
1979	-0.154					0.324			
		Add.	-0.140	-0.145	-0.158		0.127	0.161	0.223
		Con.	-0.114	-0.137	-0.106		0.076	0.144	0.107
		EDTA	-0.100	-0.064	-0.109		0.070	0.317	0.363
1980	-0.243					0.711			
		Add.	0.041	-0.001	-0.090		0.420	0.474	0.582
		Con.	0.084	-0.018	-0.030		0.458	0.638	0.635
		EDTA	-0.108	-0.014	-0.078		0.496	0.623	0.240
1981	-0.557					0.675			
		Add.	-0.521	-0.540	-0.568		0.577	0.606	0.662
		Con.	-0.420	-0.531	-0.555		0.524	0.596	0.578
		EDTA	-0.480	-0.512	-0.546		0.444	0.540	0.581
1982	-0.416					0.233			
		Add.	-0.305	-0.330	-0.373		0.296	0.294	0.287
		Con.	-0.190	-0.286	-0.301		0.334	0.288	0.266
		EDTA	-0.240	-0.292	-0.304		0.278	0.322	0.253
1983	-0.711					0.165			
		Add.	-0.517	-0.560	-0.640		0.173	0.176	0.176
		Con.	-0.407	-0.574	-0.608		0.242	0.177	0.190
		EDTA	-0.401	-0.543	-0.587		0.200	0.237	0.205

Table H8. Correlations between yield and sludge applications, cabbage and lettuce on calcareous loam soil.

	Cabbage ds		Cabbage Ni	Cabbage Cu	Cabbage Zn	Lettuce ds	Lettuce Ni	Lettuce Cu	Lettuce Zn
1979	0.437					0.719			
		Add.	0.338	0.363	0.403		0.536	0.579	0.651
		Con.	0.326	0.397	0.339		0.465	0.621	0.611
		EDTA	0.329	0.563	0.368		0.463	0.608	0.610
1980	0.177					0.246			
		Add.	0.176	0.181	0.186		0.147	0.167	0.203
		Con.	0.128	0.144	0.200		0.046	0.100	0.082
		EDTA	0.180	0.168	0.169		0.009	0.059	0.071
1981	0.184					0.564			
		Add.	0.055	0.077	0.117		0.407	0.442	0.504
		Con.	0.019	0.033	0.021		0.373	0.440	0.444
		EDTA	0.067	0.019	0.025		0.354	0.427	0.478
1982	0.419					0.163			
		Add.	0.357	0.377	0.405		0.117	0.128	0.145
		Con.	0.353	0.385	0.344		0.148	0.110	0.104
		EDTA	0.369	0.399	0.393		0.134	0.138	0.195
1983	0.684					0.159			
		Add.	0.510	0.547	0.628		0.327	0.313	0.268
		Con.	0.477	0.433	0.492		0.135	0.291	0.243
		EDTA	0.382	0.463	0.494		0.477	0.366	0.351

Table H9. Correlations between yield and sludge applications, ryegrass and red beet on calcareous loam soil.

	Ryegrass ds		Ni	Cu	Zn	Red beet ds	Ni	Cu	Zn
1979	0.812					0.524			
		Add.	0.646	0.689	0.761		0.561	0.571	0.578
		Con.	0.558	0.657	0.656		0.523	0.544	0.441
		EDTA	0.461	0.619	0.545		0.551	0.672	0.647
1980	0.178					0.314			
		Add.	0.093	0.109	0.137		0.236	0.254	0.287
		Con.	0.083	0.043	0.063		0.269	0.231	0.276
		EDTA	-0.010	0.048	0.045		0.192	0.232	0.245
1981	-0.511					0.674			
		Add.	-0.291	-0.332	-0.413		0.466	0.510	0.589
		Con.	-0.235	-0.334	-0.461		0.350	0.524	0.517
		EDTA	-0.249	-0.406	-0.382		0.437	0.526	0.556
1982	-0.448					0.115			
		Add.	-0.247	-0.283	-0.356		0.263	0.249	0.207
		Con.	-0.344	-0.378	-0.374		0.278	0.315	0.218
		EDTA	-0.206	-0.338	-0.351		0.244	0.288	0.263
1983	-0.374					0.563			
		Add.	-0.384	-0.393	-0.401		0.544	0.562	0.584
		Con.	-0.399	-0.408	-0.415		0.479	0.393	0.388
		EDTA	-0.354	-0.406	-0.419		0.394	0.366	0.360

APPENDIX I

MEAN CONCENTRATIONS OF METALS IN CROPS

Table I1. Mean concentrations of Cd in wheat

Soil	Sludge		1979	1980	1981	1982	1983	SE
sandy loam	none	mean ($\bar{x}$)	0.10	0.17	0.13	0.14	0.13	0.032
	liquid	$\bar{x}$	0.19	0.38	0.25	0.32	0.33	0.018
	bed-dried	$\bar{x}$	NA	0.29	0.35	0.33	0.36	0.029
clay	none	$\bar{x}$	0.15	0.23	0.21	0.32	0.38	0.054
	liquid	$\bar{x}$	0.32	0.54	0.63	0.43	0.70	0.031
	bed-dried	$\bar{x}$	NA	0.32	0.59	0.40	0.88	0.060
calcareous loam	none	$\bar{x}$	0.07	0.04	0.04	0.06	0.05	0.025
	liquid	$\bar{x}$	0.14	0.18	0.16	0.14	0.13	0.015
	bed-dried	$\bar{x}$	NA	0.26	0.35	0.27	0.18	0.041

Table I2. Mean concentrations of Ni in wheat

Soil	Sludge		1979	1980	1981	1982	1983	SE
sandy loam	none	mean ($\bar{x}$)	2.7	1.7	1.0	1.6	0.7	0.35
	liquid	$\bar{x}$	2.8	1.6	1.1	1.9	1.4	0.20
	bed-dried	$\bar{x}$	NA	2.0	1.0	1.7	1.1	0.27
clay	none	$\bar{x}$	4.3	1.0	1.0	0.5	1.2	0.50
	liquid	$\bar{x}$	3.8	1.1	1.1	0.6	1.7	0.29
	bed-dried	$\bar{x}$	NA	1.3	2.3	0.5	1.6	0.41
calcareous loam	none	$\bar{x}$	0.3	0.5	0.6	1.5	1.0	0.24
	liquid	$\bar{x}$	0.4	0.9	0.6	1.8	1.0	0.14
	bed-dried	$\bar{x}$	NA	0.6	0.7	2.0	1.0	0.17

Table I3. Mean concentrations of Cu in wheat

Soil	Sludge		1979	1980	1981	1982	1983	SE
sandy loam	none	mean ($\bar{x}$)	3.9	6.0	3.5	7.9	3.8	0.49
	liquid	$\bar{x}$	6.2	7.8	4.7	8.2	5.7	0.28
	bed-dried	$\bar{x}$	NA	10.3	5.5	10.1	7.0	0.81
clay	none	$\bar{x}$	4.8	6.3	4.7	9.1	4.8	0.73
	liquid	$\bar{x}$	5.0	6.4	5.7	9.7	6.4	0.42
	bed-dried	$\bar{x}$	NA	7.8	6.0	9.2	7.8	0.73
calcareous loam	none	$\bar{x}$	4.4	5.0	6.0	5.5	5.1	0.24
	liquid	$\bar{x}$	4.8	5.6	6.3	6.7	5.4	0.14
	bed-dried	$\bar{x}$	NA	6.8	7.2	7.7	5.5	0.32

Table I4. Mean concentrations of Zn in wheat

Soil	Sludge		1979	1980	1981	1982	1983	SE
sandy loam	none	mean ($\bar{x}$)	26.0	40.7	20.2	34.2	23.8	4.35
	liquid	$\bar{x}$	52.7	69.5	49.1	64.4	58.0	2.51
	bed-dried	$\bar{x}$	NA	69.8	47.8	74.3	55.8	4.14
clay	none	$\bar{x}$	38.0	34.5	21.5	30.3	37.6	4.46
	liquid	$\bar{x}$	45.3	56.7	51.1	45.4	70.7	2.58
	bed-dried	$\bar{x}$	NA	64.5	45.3	39.5	71.3	3.45
calcareous loam	none	$\bar{x}$	29.1	45.0	40.5	35.7	37.2	2.40
	liquid	$\bar{x}$	35.3	60.8	47.6	55.8	45.9	1.38
	bed-dried	$\bar{x}$	NA	72.5	55.5	67.5	48.5	3.67

Table I5. Mean concentrations of Pb in wheat

Soil	Sludge		1979	1980	1981	1982	1983	SE
sandy loam	none	mean ($\bar{x}$)	0.7	1.3	1.0	0.5	0.5	0.08
	liquid	$\bar{x}$	0.8	1.1	1.0	0.5	0.5	0.04
	bed-dried	$\bar{x}$	NA	1.0	1.0	0.7	0.5	0.11
clay	none	$\bar{x}$	0.5	1.0	1.0	0.5	1.0	0.10
	liquid	$\bar{x}$	0.6	1.0	1.0	0.5	1.0	0.06
	bed-dried	$\bar{x}$	NA	1.0	1.0	0.5	1.3	0.06
calcareous loam	none	$\bar{x}$	0.8	0.6	0.6	1.0	1.2	0.11
	liquid	$\bar{x}$	0.7	0.6	0.6	1.2	1.1	0.06
	bed-dried	$\bar{x}$	NA	0.6	0.6	1.0	1.0	0.08

Table I6. Mean concentrations of Cd in potatoes

Soil	Sludge		1979	1980	1981	1982	1983	SE
sandy loam	none	mean ($\bar{x}$)	0.10	0.13	0.19	0.16	0.16	0.022
	liquid	$\bar{x}$	0.19	0.19	0.20	0.20	0.16	0.012
	bed-dried	$\bar{x}$	NA	0.17	0.26	0.21	0.16	0.028
clay	none	$\bar{x}$	0.50	0.44	0.50	0.46	0.70	0.039
	liquid	$\bar{x}$	0.58	0.38	0.47	0.39	0.63	0.023
	bed-dried	$\bar{x}$	NA	0.26	0.36	0.31	0.69	0.049
calcareous loam	none	$\bar{x}$	0.09	0.11	0.04	0.05	0.03	0.017
	liquid	$\bar{x}$	0.13	0.15	0.08	0.14	0.08	0.010
	bed-dried	$\bar{x}$	NA	0.19	0.10	0.16	0.13	0.021

Table I7. Mean concentrations of Ni in potatoes

Soil	Sludge		1979	1980	1981	1982	1983	SE
sandy loam	none	mean ($\bar{x}$)	0.9	1.0	1.7	1.6	2.3	0.27
	liquid	$\bar{x}$	1.8	1.3	1.8	1.5	2.3	0.16
	bed-dried	$\bar{x}$	NA	1.0	2.0	1.8	1.8	0.32
clay	none	$\bar{x}$	1.9	1.0	1.0	1.3	0.7	0.18
	liquid	$\bar{x}$	2.4	1.7	1.4	1.6	1.4	0.10
	bed-dried	$\bar{x}$	NA	1.0	1.0	1.9	1.7	0.12
calcareous loam	none	$\bar{x}$	0.4	0.1	0.8	1.2	0.6	0.09
	liquid	$\bar{x}$	0.5	0.1	0.8	1.1	0.6	0.05
	bed-dried	$\bar{x}$	NA	0.1	0.8	1.5	0.7	0.13

Table I8. Mean concentrations of Cu in potatoes

Soil	Sludge		1979	1980	1981	1982	1983	SE
sandy loam	none	mean ($\bar{x}$)	5.0	4.5	6.8	6.6	6.8	0.48
	liquid	$\bar{x}$	8.8	7.1	7.2	8.2	7.7	0.27
	bed-dried	$\bar{x}$	NA	9.8	10.8	9.1	9.6	0.55
clay	none	$\bar{x}$	8.3	8.5	7.2	9.0	8.3	0.28
	liquid	$\bar{x}$	10.4	9.5	8.3	9.7	8.7	0.16
	bed-dried	$\bar{x}$	NA	10.3	9.5	10.0	9.3	0.37
calcareous loam	none	$\bar{x}$	4.9	7.3	6.8	7.9	9.2	0.42
	liquid	$\bar{x}$	7.9	9.6	9.2	9.9	10.6	0.24
	bed-dried	$\bar{x}$	NA	12.8	11.2	11.1	11.0	0.45

Table I9. Mean concentrations of Zn in potatoes

Soil	Sludge		1979	1980	1981	1982	1983	SE
sandy loam	none	mean ($\bar{x}$)	14.3	10.7	14.7	12.7	13.2	0.94
	liquid	$\bar{x}$	19.8	15.0	16.6	15.3	15.2	0.54
	bed-dried	$\bar{x}$	NA	17.3	17.0	15.5	15.3	0.75
clay	none	$\bar{x}$	17.7	15.3	11.7	14.3	17.7	0.93
	liquid	$\bar{x}$	20.4	18.9	16.8	16.8	18.7	0.54
	bed-dried	$\bar{x}$	NA	17.0	16.5	16.5	18.8	1.02
calcareous loam	none	$\bar{x}$	11.0	14.5	15.1	17.7	17.8	1.07
	liquid	$\bar{x}$	16.6	16.3	17.4	17.8	18.1	0.62
	bed-dried	$\bar{x}$	NA	19.1	16.1	17.3	18.3	1.03

Table I10. Mean concentrations of Pb in potatoes
Results mainly on or below the limit of detection; 0.5-1 mg/kg

Table I11. Mean concentrations of Cd in lettuce

Soil	Sludge		1979	1980	1981	1982	1983	SE
sandy loam	none	mean ($\bar{x}$)	0.95	0.76	1.12	1.03	1.14	0.247
	liquid	$\bar{x}$	1.88	2.49	2.74	2.51	2.16	0.143
	bed-dried	$\bar{x}$	NA	2.50	1.95	2.48	2.15	0.173
clay	none	$\bar{x}$	4.77	3.02	5.10	5.23	5.24	1.533
	liquid	$\bar{x}$	6.91	4.72	5.22	6.42	13.65	0.885
	bed-dried	$\bar{x}$	NA	3.90	4.93	5.47	8.66	0.701
calcareous loam	none	$\bar{x}$	0.68	0.29	0.57	0.40	1.23	0.163
	liquid	$\bar{x}$	1.63	1.12	0.91	0.87	1.91	0.094
	bed-dried	$\bar{x}$	NA	2.26	1.60	1.55	2.55	0.284

Table I12. Mean concentrations of Ni in lettuce

Soil	Sludge		1979	1980	1981	1982	1983	SE
sandy loam	none	mean ($\bar{x}$)	7.4	6.0	9.8	3.5	3.5	0.87
	liquid	$\bar{x}$	7.1	6.9	10.7	6.4	5.0	0.50
	bed-dried	$\bar{x}$	NA	8.3	5.5	4.3	4.0	0.80
clay	none	$\bar{x}$	6.0	2.8	3.5	3.8	5.1	0.61
	liquid	$\bar{x}$	6.5	4.7	5.0	6.1	8.5	0.35
	bed-dried	$\bar{x}$	NA	5.0	5.0	4.0	6.0	0.74
calcareous loam	none	$\bar{x}$	0.7	0.6	0.9	1.8	2.0	0.25
	liquid	$\bar{x}$	1.4	0.9	0.9	2.1	2.2	0.14
	bed-dried	$\bar{x}$	NA	1.8	1.3	2.8	4.0	0.38

Table I13. Mean concentrations of Cu in lettuce

Soil	Sludge		1979	1980	1981	1982	1983	SE
sandy loam	none	mean ($\bar{x}$)	11.2	11.2	13.8	10.5	8.3	0.85
	liquid	$\bar{x}$	17.1	17.8	19.7	14.8	11.8	0.49
	bed-dried	$\bar{x}$	NA	21.0	21.8	19.5	15.5	0.89
clay	none	$\bar{x}$	24.0	16.8	15.5	15.3	19.2	1.08
	liquid	$\bar{x}$	23.2	17.0	14.3	14.3	20.6	0.62
	bed-dried	$\bar{x}$	NA	15.3	14.3	17.7	18.3	1.02
calcareous loam	none	$\bar{x}$	12.7	9.4	17.2	11.8	14.8	0.79
	liquid	$\bar{x}$	14.7	12.8	17.5	10.5	14.8	0.46
	bed-dried	$\bar{x}$	NA	17.1	19.6	12.6	15.5	0.93

Table I14. Mean concentrations of Zn in lettuce

Soil	Sludge		1979	1980	1981	1982	1983	SE
sandy loam	none	mean ($\bar{x}$)	69.3	61.0	66.8	51.3	47.2	5.18
	liquid	$\bar{x}$	117.6	110.2	115.4	91.4	79.8	2.99
	bed-dried	$\bar{x}$	NA	101.0	100.0	86.3	77.5	3.89
clay	none	$\bar{x}$	99.0	67.7	71.2	76.2	84.8	13.77
	liquid	$\bar{x}$	152.5	101.7	109.7	130.9	180.6	
7.95								
	bed-dried	$\bar{x}$	NA	82.3	96.8	109.8	133.8	6.04
calcareous loam	none	$\bar{x}$	50.8	53.7	72.7	64.5	78.7	4.00
	liquid	$\bar{x}$	75.9	72.1	76.8	67.4	78.0	2.31
	bed-dried	$\bar{x}$	NA	86.5	76.5	76.3	80.0	4.73

Table I15. Mean concentrations of Pb in lettuce

Soil	Sludge		1979	1980	1981	1982	1983	SE
sandy loam	none	mean ($\bar{x}$)	2.8	1.0	1.2	0.6	0.8	0.34
	liquid	$\bar{x}$	4.1	1.1	1.1	0.7	0.8	0.20
	bed-dried	$\bar{x}$	NA	1.0	1.0	0.8	0.9	0.14
clay	none	$\bar{x}$	4.7	1.0	1.3	0.7	3.7	0.53
	liquid	$\bar{x}$	3.2	1.1	1.3	0.8	5.3	0.30
	bed-dried	$\bar{x}$	NA	1.0	1.8	0.8	7.3	1.19
calcareous loam	none	$\bar{x}$	5.6	3.5	3.6	2.3	4.3	0.58
	liquid	$\bar{x}$	4.9	2.6	2.6	2.6	4.6	0.33
	bed-dried	$\bar{x}$	NA	3.0	3.2	3.5	3.7	0.75

Table I16. Mean concentrations of Cd in cabbage

Soil	Sludge		1979	1980	1981	1982	1983	SE
sandy loam	none	mean ($\bar{x}$)	0.10	0.12	0.10	0.13	0.12	0.061
	liquid	$\bar{x}$	0.10	0.17	0.13	0.16	0.20	0.009
	bed-dried	$\bar{x}$	NA	0.27	0.20	0.18	0.27	0.021
clay	none	$\bar{x}$	0.35	0.33	0.24	0.24	0.17	0.024
	liquid	$\bar{x}$	0.33	0.40	0.27	0.22	0.18	0.014
	bed-dried	$\bar{x}$	NA	0.42	0.33	0.19	0.20	0.031
calcareous loam	none	$\bar{x}$	0.07	0.06	0.06	0.06	0.02	0.025
	liquid	$\bar{x}$	0.26	0.15	0.11	0.12	0.10	0.014
	bed-dried	$\bar{x}$	NA	0.67	0.42	0.23	0.23	0.057

Table I17. Mean concentrations of Ni in cabbage

Soil	Sludge		1979	1980	1981	1982	1983	SE
sandy loam	none	mean ($\bar{x}$)	4.2	7.8	1.7	1.4	4.7	0.67
	liquid	$\bar{x}$	4.5	6.7	4.2	3.6	4.7	0.39
	bed-dried	$\bar{x}$	NA	3.8	3.5	2.0	5.0	0.81
clay	none	$\bar{x}$	1.6	2.2	2.8	1.8	1.4	0.43
	liquid	$\bar{x}$	3.2	4.3	4.1	4.4	3.1	0.25
	bed-dried	$\bar{x}$	NA	5.8	3.0	3.3	2.9	0.34
calcareous loam	none	$\bar{x}$	0.8	0.6	0.6	1.7	1.0	0.22
	liquid	$\bar{x}$	2.3	1.2	0.9	2.4	1.2	0.13
	bed-dried	$\bar{x}$	NA	4.9	2.2	3.8	1.8	0.37

Table I18. Mean concentrations of Cu in cabbage

Soil	Sludge		1979	1980	1981	1982	1983	SE
sandy loam	none	mean ($\bar{x}$)	3.4	3.7	3.0	3.4	5.0	0.26
	liquid	$\bar{x}$	5.6	5.4	3.8	4.4	5.1	0.15
	bed-dried	$\bar{x}$	NA	6.8	4.8	5.1	6.7	0.27
clay	none	$\bar{x}$	3.6	3.3	3.0	3.3	3.3	0.25
	liquid	$\bar{x}$	5.6	5.5	4.3	4.8	3.9	0.14
	bed-dried	$\bar{x}$	NA	6.5	4.8	4.5	4.5	0.29
calcareous loam	none	$\bar{x}$	2.9	2.1	3.4	2.6	3.0	0.22
	liquid	$\bar{x}$	4.3	3.5	4.0	3.1	4.2	0.13
	bed-dried	$\bar{x}$	NA	7.2	6.5	4.3	5.3	0.32

Table I19. Mean concentrations of Zn in cabbage

Soil	Sludge		1979	1980	1981	1982	1983	SE
sandy loam	none	mean ($\bar{x}$)	20.7	26.7	20.0	20.7	36.0	1.93
	liquid	$\bar{x}$	32.4	43.8	29.7	35.1	41.3	1.11
	bed-dried	$\bar{x}$	NA	46.5	32.0	35.8	39.0	1.96
clay	none	$\bar{x}$	31.5	23.5	19.3	21.3	20.5	2.40
	liquid	$\bar{x}$	44.9	50.9	37.9	35.6	34.4	1.39
	bed-dried	$\bar{x}$	NA	48.5	34.5	33.0	34.3	1.73
calcareous loam	none	$\bar{x}$	26.6	26.7	23.8	22.8	25.4	2.10
	liquid	$\bar{x}$	41.4	29.1	29.8	25.6	34.1	1.22
	bed-dried	$\bar{x}$	NA	74.5	47.9	32.4	37.1	3.32

Table I20. Mean concentrations of Pb in cabbage

Soil	Sludge		1979	1980	1981	1982	1983	SE
sandy loam	none	mean ($\bar{x}$)	0.5	1.0	1.0	0.5	0.5	0.14
	liquid	$\bar{x}$	1.2	1.0	1.1	0.5	0.5	0.08
	bed-dried	$\bar{x}$	NA	1.0	1.0	0.5	0.5	0.02
clay	none	$\bar{x}$	0.5	1.0	1.0	0.5	0.6	0.02
	liquid	$\bar{x}$	0.5	1.0	1.0	0.5	0.5	0.01
	bed-dried	$\bar{x}$	NA	1.0	1.0	0.5	0.5	0.03
calcareous loam	none	$\bar{x}$	1.0	0.7	0.9	1.5	1.0	0.16
	liquid	$\bar{x}$	1.1	0.7	1.0	1.6	1.1	0.09
	bed-dried	$\bar{x}$	NA	0.9	0.9	1.5	1.0	0.18

Table I21. Mean concentrations of Cd in red beet

Soil	Sludge		1979	1980	1981	1982	1983	SE
sandy loam	none	mean ($\bar{x}$)	0.50	0.23	0.16	0.28	0.26	0.084
	liquid	$\bar{x}$	0.98	0.67	0.50	0.56	0.44	0.048
	bed-dried	$\bar{x}$	NA	0.78	0.69	0.79	0.51	0.064
clay	none	$\bar{x}$	1.07	0.92	0.77	0.77	0.74	0.117
	liquid	$\bar{x}$	1.56	1.47	1.08	1.04	0.97	0.068
	bed-dried	$\bar{x}$	NA	2.12	1.06	0.97	0.90	0.127
calcareous loam	none	$\bar{x}$	0.11	0.12	0.13	0.09	0.07	0.030
	liquid	$\bar{x}$	0.27	0.27	0.33	0.21	0.20	0.017
	bed-dried	$\bar{x}$	NA	0.62	0.53	0.36	0.45	0.041

Table I22. Mean concentrations of Ni in red beet

Soil	Sludge		1979	1980	1981	1982	1983	SE
sandy loam	none	mean ($\bar{x}$)	7.1	1.5	4.2	1.9	2.5	1.06
	liquid	$\bar{x}$	6.8	3.7	2.2	3.6	3.0	0.61
	bed-dried	$\bar{x}$	NA	3.0	1.8	2.9	3.2	1.61
clay	none	$\bar{x}$	1.5	2.3	1.5	1.8	0.7	0.52
	liquid	$\bar{x}$	2.8	4.9	2.8	3.3	2.1	0.30
	bed-dried	$\bar{x}$	NA	2.8	1.5	1.3	2.4	0.45
calcareous loam	none	$\bar{x}$	0.6	0.6	0.8	1.0	2.0	0.07
	liquid	$\bar{x}$	0.7	0.5	0.8	1.0	2.0	0.04
	bed-dried	$\bar{x}$	NA	0.6	0.7	1.0	2.0	0.02

Table I23. Mean concentrations of Cu in red beet

Soil	Sludge		1979	1980	1981	1982	1983	SE
sandy loam	none	mean ($\bar{x}$)	8.3	6.7	6.5	8.4	7.1	0.32
	liquid	$\bar{x}$	9.3	8.7	7.4	9.3	7.3	0.18
	bed-dried	$\bar{x}$	NA	10.0	9.0	9.7	6.8	0.53
clay	none	$\bar{x}$	9.6	7.2	7.3	10.8	9.2	0.74
	liquid	$\bar{x}$	10.0	7.7	7.8	9.6	9.2	0.43
	bed-dried	$\bar{x}$	NA	9.5	7.5	8.8	9.3	1.35
calcareous loam	none	$\bar{x}$	9.6	8.4	7.0	6.8	6.2	0.38
	liquid	$\bar{x}$	9.8	9.1	8.5	7.9	7.6	0.22
	bed-dried	$\bar{x}$	NA	10.4	9.2	9.4	8.4	0.53

Table I24. Mean concentrations of Zn in red beet

Soil	Sludge		1979	1980	1981	1982	1983	SE
sandy loam	none	mean ($\bar{x}$)	32.3	24.3	22.2	27.3	26.8	5.28
	liquid	$\bar{x}$	66.4	64.1	55.5	57.3	43.3	3.05
	bed-dried	$\bar{x}$	NA	38.9	47.8	48.5	36.5	2.46
clay	none	$\bar{x}$	36.3	34.5	26.5	28.0	30.7	6.00
	liquid	$\bar{x}$	71.9	102.3	71.4	62.2	55.9	3.46
	bed-dried	$\bar{x}$	NA	70.5	48.3	45.0	45.5	4.28
calcareous loam	none	$\bar{x}$	29.2	31.6	22.4	23.9	20.8	1.50
	liquid	$\bar{x}$	35.7	34.5	31.8	29.6	26.5	0.87
	bed-dried	$\bar{x}$	NA	30.3	34.5	32.8	30.0	1.79

Table I25. Mean concentrations of Pb in red beet

Soil	Sludge		1979	1980	1981	1982	1983	SE
sandy loam	none	mean ($\bar{x}$)	0.5	1.0	1.0	0.5	0.5	0.09
	liquid	$\bar{x}$	0.5	1.0	1.0	1.7	0.5	0.05
	bed-dried	$\bar{x}$	NA	1.0	1.0	1.1	0.4	0.15
clay	none	$\bar{x}$	0.5	1.0	1.0	0.6	0.5	0.07
	liquid	$\bar{x}$	0.6	1.0	1.0	0.6	0.6	0.04
	bed-dried	$\bar{x}$	NA	1.0	1.0	0.6	0.6	0.04
calcareous loam	none	$\bar{x}$	1.3	1.0	0.8	1.0	1.5	0.14
	liquid	$\bar{x}$	1.3	1.2	0.8	1.0	1.3	0.08
	bed-dried	$\bar{x}$	NA	1.0	1.0	1.0	1.3	0.15

Table I26. Mean concentrations of Cd in ryegrass

Soil	Sludge		1979	1980	1981	1982	1983	SE
sandy loam	none	mean ($\bar{x}$)	0.10	0.10	0.10	0.13	0.11	0.033
	liquid	$\bar{x}$	0.38	0.29	0.28	0.30	0.26	0.019
	bed-dried	$\bar{x}$	NA	0.45	0.20	0.26	0.22	0.052
clay	none	$\bar{x}$	0.26	0.18	0.22	0.13	0.16	0.037
	liquid	$\bar{x}$	0.59	0.43	0.56	0.41	0.39	0.022
	bed-dried	$\bar{x}$	NA	0.69	0.48	0.39	0.35	0.042
calcareous loam	none	$\bar{x}$	0.02	0.04	0.05	0.10	0.01	0.014
	liquid	$\bar{x}$	0.09	0.14	0.18	0.18	0.04	0.008
	bed-dried	$\bar{x}$	NA	0.47	0.47	0.32	0.15	0.041

Table I27. Mean concentrations of Ni in ryegrass

Soil	Sludge		1979	1980	1981	1982	1983	SE
sandy loam	none	mean ($\bar{x}$)	3.4	3.1	3.0	3.3	3.2	0.45
	liquid	$\bar{x}$	7.3	8.1	7.6	7.6	7.6	0.26
	bed-dried	$\bar{x}$	NA	14.5	5.0	5.1	5.3	1.00
clay	none	$\bar{x}$	4.5	4.7	3.5	3.5	3.0	0.53
	liquid	$\bar{x}$	7.3	7.7	8.0	8.9	6.7	0.31
	bed-dried	$\bar{x}$	NA	11.3	6.5	6.8	6.0	0.79
calcareous loam	none	$\bar{x}$	2.3	1.3	1.3	2.2	1.1	0.35
	liquid	$\bar{x}$	6.1	3.7	3.6	3.7	2.3	0.20
	bed-dried	$\bar{x}$	NA	21.4	15.1	12.4	7.0	1.48

Table I28. Mean concentrations of Cu in ryegrass

Soil	Sludge		1979	1980	1981	1982	1983	SE
sandy loam	none	mean ($\bar{x}$)	6.6	6.0	6.0	6.9	6.7	0.49
	liquid	$\bar{x}$	14.3	11.4	11.0	11.2	11.4	0.28
	bed-dried	$\bar{x}$	NA	26.0	12.0	11.4	12.3	1.03
clay	none	$\bar{x}$	9.5	9.3	8.2	8.0	7.2	0.46
	liquid	$\bar{x}$	13.9	11.8	11.5	12.0	10.9	0.27
	bed-dried	$\bar{x}$	NA	18.3	12.5	11.8	11.5	0.65
calcareous loam	none	$\bar{x}$	10.8	8.9	10.4	10.2	7.5	0.34
	liquid	$\bar{x}$	14.8	11.3	13.5	12.4	9.4	0.20
	bed-dried	$\bar{x}$	NA	21.5	17.9	15.4	12.0	0.76

Table I29. Mean concentrations of Zn in ryegrass

Soil	Sludge		1979	1980	1981	1982	1983	SE
sandy loam	none	mean ($\bar{x}$)	27.8	25.6	22.2	25.4	23.7	2.94
	liquid	$\bar{x}$	70.7	79.3	64.6	64.8	61.8	1.70
	bed-dried	$\bar{x}$	NA	79.0	48.5	49.5	53.5	2.90
clay	none	$\bar{x}$	34.0	34.6	25.8	24.8	26.4	2.75
	liquid	$\bar{x}$	64.5	78.1	72.6	70.9	66.3	1.59
	bed-dried	$\bar{x}$	NA	71.1	53.3	54.0	56.3	2.35
calcareous loam	none	$\bar{x}$	36.8	34.4	36.7	36.7	30.1	1.65
	liquid	$\bar{x}$	59.4	55.8	57.4	54.5	43.5	0.95
	bed-dried	$\bar{x}$	NA	90.8	82.9	68.7	50.9	4.36

Table I30. Mean concentrations of Pb in ryegrass

Soil	Sludge		1979	1980	1981	1982	1983	SE
sandy loam	none	mean ($\bar{x}$)	2.9	2.0	1.5	1.8	1.6	0.19
	liquid	$\bar{x}$	3.3	2.1	1.5	1.8	2.1	0.11
	bed-dried	$\bar{x}$	NA	2.3	1.6	1.8	2.3	0.14
clay	none	$\bar{x}$	3.0	2.7	1.8	2.2	2.1	0.14
	liquid	$\bar{x}$	3.3	2.6	1.7	2.5	2.3	0.08
	bed-dried	$\bar{x}$	NA	4.0	2.0	2.3	2.3	0.19
calcareous loam	none	$\bar{x}$	4.5	3.4	3.9	3.4	2.4	0.22
	liquid	$\bar{x}$	3.6	3.4	4.1	3.5	3.5	0.13
	bed-dried	$\bar{x}$	NA	5.4	3.9	3.0	3.0	0.39

APPENDIX J

CORRELATIONS BETWEEN CROP METAL AND SOIL METALS

Table J1. Correlations for relation between crop metal and soil metal; wheat on sandy loam.

	correlation coefficient r					percent variance V%				
	Cd	Ni	Cu	Zn	Pb	Cd	Ni	Cu	Zn	Pb
1979										
a	0.836	NS	0.828	0.844	NS	69	-	67	70	-
La	0.752	NS	0.835	0.814	NS	55	-	69	65	-
c	0.845	NS	0.803	0.806	NS	70	-	63	63	-
Lc	0.734	NS	0.837	0.833	NS	52	-	69	68	-
e	0.820	NS	0.799	0.797	NS	66	-	62	62	-
Le	0.737	NS	0.834	0.838	NS	52	-	68	69	-
1980										
a	0.838	NS	0.686	0.744	NS	69	-	45	54	-
La	0.808	NS	0.569	0.668	NS	64	-	30	42	-
c	0.820	NS	0.665	0.689	NS	66	-	42	45	-
Lc	0.741	NS	0.662	0.717	NS	53	-	41	49	-
e	0.788	NS	0.628	0.703	NS	61	-	37	47	-
Le	0.746	NS	0.583	0.707	NS	54	-	31	48	-
1981										
a	0.897	0.560	0.683	0.857	NS	80	29	44	72	-
La	0.809	NS	0.676	0.911	NS	64	-	43	82	-
c	0.887	0.426	0.725	0.857	NS	78	15	51	72	-
Lc	0.782	0.401	0.751	0.898	NS	59	13	55	80	-
e	0.868	0.413	0.685	0.828	NS	74	14	45	67	-
Le	0.789	NS	0.727	0.918	NS	61	-	51	84	-
1982										
a	0.885	0.623	NS	0.768	NS	78	36	-	57	-
La	0.864	0.420	NS	0.656	NS	74	14	-	41	-
c	0.918	0.395	NS	0.812	NS	84	12	-	65	-
Lc	0.854	NS	NS	0.800	NS	72	-	-	63	-
e	0.921	0.503	NS	0.848	NS	84	22	-	71	-
Le	0.839	0.454	NS	0.806	NS	69	17	-	63	-
1983										
a	0.868	0.620	0.879	0.928	NS	74	36	76	86	-
La	0.820	0.564	0.752	0.803	NS	66	29	55	63	-
c	0.893	0.471	0.851	0.864	NS	79	19	71	74	-
Lc	0.828	0.462	0.865	0.897	NS	67	18	74	80	-
e	0.907	0.628	0.842	0.853	NS	81	37	70	72	-
Le	0.832	0.618	0.846	0.856	NS	68	36	70	72	-

Table J2. Correlations for relation between crop metal and soil metal; wheat on clay.

	correlation coefficient r					percent variance V%				
	Cd	Ni	Cu	Zn	Pb	Cd	Ni	Cu	Zn	Pb
1979										
a	0.718	NS	0.427	0.654	NS	49	-	15	40	-
La	0.673	NS	NS	0.523	NS	43	-	-	24	-
c	0.746	NS	NS	0.619	NS	54	-	-	36	-
Lc	0.695	NS	NS	0.621	NS	46	-	-	36	-
e	0.749	NS	NS	0.638	NS	54	-	-	38	-
Le	0.701	NS	NS	0.560	NS	47	-	-	28	-
1980										
a	0.876	0.540	0.487	0.874	NS	76	26	21	75	-
La	0.836	NS	NS	0.776	NS	69	-	-	59	-
c	0.816	NS	0.426	0.814	NS	65	-	15	65	-
Lc	0.768	NS	NS	0.825	NS	57	-	-	67	-
e	0.805	0.490	0.438	0.842	NS	63	21	16	70	-
Le	0.769	0.424	NS	0.820	NS	57	15	-	66	-
1981										
a	0.903	0.560	0.462	0.908	NS	81	29	18	82	-
La	0.904	NS	0.541	0.884	NS	81	-	26	77	-
c	0.841	0.598	0.421	0.830	NS	70	33	14	68	-
Lc	0.890	0.568	0.491	0.916	NS	78	29	21	83	-
e	0.884	0.614	0.430	0.857	NS	77	35	15	72	-
Le	0.902	0.472	0.503	0.940	NS	81	19	22	88	-
1982										
a	0.898	0.890	0.632	0.819	NS	80	78	34	66	-
La	0.790	0.552	NS	0.672	NS	61	28	-	43	-
c	0.808	0.784	0.507	0.788	NS	64	60	22	61	-
Lc	0.691	0.739	0.449	0.797	NS	46	53	16	62	-
e	0.824	0.731	0.574	0.768	NS	67	51	30	57	-
Le	0.726	0.651	0.438	0.744	NS	51	40	15	54	-
1983										
a	0.650	0.617	0.710	0.623	NS	40	35	48	36	-
La	0.669	0.502	0.573	0.599	NS	43	22	30	33	-
c	0.727	0.605	0.784	0.671	NS	51	34	60	43	-
Lc	0.674	0.586	0.737	0.667	NS	43	32	52	42	-
e	0.754	0.655	0.792	0.742	NS	55	40	61	53	-
Le	0.693	0.607	0.723	0.724	NS	46	34	50	50	-

Table J3. Correlations for relation between crop metal and soil metal; wheat on calcareous loam.

	correlation coefficient r					percent variance V%				
	Cd	Ni	Cu	Zn	Pb	Cd	Ni	Cu	Zn	Pb
1979										
a	0.649	0.402	0.484	0.633	NS	40	13	20	38	-
La	0.657	NS	0.411	0.617	NS	41	-	13	36	-
c	0.610	NS	0.465	0.478	NS	35	-	18	20	-
Lc	0.576	NS	0.429	0.484	NS	30	-	15	20	-
e	0.645	0.461	0.450	0.562	NS	39	18	17	29	-
Le	0.597	0.411	NS	0.578	NS	33	13	-	31	-
1980										
a	0.889	NS	0.547	0.800	NS	78	-	27	62	-
La	0.818	NS	0.655	0.751	NS	66	-	41	55	-
c	0.862	NS	NS	0.821	NS	73	-	-	66	-
Lc	0.793	NS	0.510	0.848	NS	61	-	23	71	-
e	0.832	NS	0.558	0.841	NS	68	-	28	69	-
Le	0.754	NS	0.653	0.854	NS	55	-	40	72	-
1981										
a	0.791	NS	NS	0.510	NS	61	-	-	23	-
La	0.842	NS	NS	0.506	NS	70	-	-	23	-
c	0.817	NS	NS	0.685	NS	65	-	-	45	-
Lc	0.853	NS	0.399	0.760	NS	72	-	12	56	-
e	0.839	NS	NS	0.667	NS	69	-	-	42	-
Le	0.868	NS	NS	0.759	NS	74	-	-	56	-
1982										
a	0.882	NS	0.580	0.902	NS	77	-	31	81	-
La	0.756	NS	0.542	0.741	NS	55	-	26	53	-
c	0.889	NS	0.613	0.868	NS	78	-	35	74	-
Lc	0.754	NS	0.602	0.854	NS	55	-	34	72	-
e	0.885	NS	0.578	0.860	NS	77	-	31	73	-
Le	0.724	NS	0.594	0.829	NS	50	-	33	67	-
1983										
a	0.563	NS	NS	0.790	NS	29	-	-	61	-
La	0.526	NS	0.411	0.640	NS	25	-	13	38	-
c	0.539	NS	NS	0.819	NS	26	-	-	66	-
Lc	0.555	NS	0.391	0.817	NS	28	-	12	65	-
e	0.488	NS	NS	0.847	NS	21	-	-	71	-
Le	0.492	NS	0.469	0.844	NS	21	-	19	70	-

Table J4. Correlations for relation between crop metal and soil metal; potato on sandy loam.

	correlation coefficient r					percent variance V%				
	Cd	Ni	Cu	Zn	Pb	Cd	Ni	Cu	Zn	Pb
1979										
a	0.646	0.775	0.870	0.717	NS	39	58	75	49	-
La	0.699	0.724	0.826	0.634	NS	47	51	67	38	-
c	0.654	0.763	0.872	0.714	NS	40	56	75	49	-
Lc	0.706	0.778	0.916	0.745	NS	48	59	83	54	-
e	0.707	0.833	0.880	0.757	NS	48	68	77	56	-
Le	0.744	0.856	0.903	0.707	NS	54	72	81	48	-
1980										
a	0.651	0.812	0.783	0.765	NS	40	64	60	57	-
Le	0.688	0.608	0.718	0.709	NS	45	34	50	48	-
c	0.660	0.760	0.758	0.818	NS	41	56	56	66	-
Lc	0.653	0.731	0.763	0.832	NS	40	52	56	68	-
e	0.671	0.763	0.730	0.799	NS	43	56	51	62	-
Le	0.663	0.695	0.729	0.785	NS	42	46	51	60	-
1981										
a	NS	NS	NS	0.409	NS	-	-	-	13	-
La	NS	NS	NS	NS	NS	-	-	-	-	-
c	NS	NS	NS	NS	NS	-	-	-	-	-
Lc	NS	NS	NS	NS	NS	-	-	-	-	-
e	NS	NS	NS	NS	NS	-	-	-	-	-
Le	NS	NS	NS	NS	NS	-	-	-	-	-
1982										
a	0.445	NS	0.553	0.575	NS	16	-	28	30	-
La	NS	NS	0.658	0.620	NS	-	-	41	36	-
c	0.511	NS	0.584	0.516	NS	23	-	31	24	-
Lc	NS	NS	0.620	0.528	NS	-	-	36	25	-
e	0.507	NS	0.584	0.528	NS	23	-	31	25	-
Le	NS	NS	0.650	0.626	NS	-	-	40	37	-
1983										
a	0.629	NS	0.392	0.616	NS	37	-	12	35	-
La	0.452	NS	NS	0.524	NS	17	-	-	24	-
c	0.619	NS	0.547	0.557	NS	36	-	27	28	-
Lc	0.397	NS	0.554	0.549	NS	12	-	28	27	-
e	0.620	NS	0.552	0.551	NS	36	-	28	27	-
Le	0.411	NS	0.550	0.559	NS	13	-	27	28	-

Table J5. Correlations for relation between crop metal and soil metal; potato on clay.

	correlation coefficient r					percent variance V%				
	Cd	Ni	Cu	Zn	Pb	Cd	Ni	Cu	Zn	Pb
1979										
a	NS	0.618	0.700	0.450	NS	-	36	47	17	-
La	NS	0.462	0.785	0.416	NS	-	18	60	14	-
c	NS	0.654	0.649	0.438	NS	-	40	40	16	-
Lc	NS	0.663	0.748	0.454	NS	-	42	54	17	-
e	NS	0.651	0.670	0.425	NS	-	40	43	15	-
Le	NS	0.619	0.759	0.416	NS	-	36	56	14	-
1980										
a	NS	0.807	NS	0.511	NS	-	64	-	23	-
La	NS	0.662	0.434	0.588	NS	-	41	15	32	-
c	NS	0.547	NS	NS	NS	-	27	-	-	-
Lc	NS	0.544	NS	0.450	NS	-	27	-	17	-
e	NS	0.804	NS	0.457	NS	-	63	-	18	-
Le	NS	0.773	NS	0.604	NS	-	58	-	34	-
1981										
a	NS	0.777	0.714	0.553	NS	-	59	49	28	-
La	NS	0.619	0.649	0.617	NS	-	36	40	36	-
c	NS	0.680	0.714	0.523	NS	-	44	49	24	-
Lc	NS	0.672	0.709	0.540	NS	-	43	48	26	-
e	NS	0.632	0.721	0.544	NS	-	37	50	27	-
Le	NS	0.628	0.712	0.598	NS	-	37	49	33	-
1982										
a	NS	0.736	0.499	0.497	NS	-	52	22	22	-
La	NS	0.481	0.444	0.389	NS	-	20	16	12	-
c	NS	0.790	0.586	0.561	NS	-	61	32	29	-
Lc	NS	0.788	0.586	0.574	NS	-	61	32	30	-
e	NS	0.734	0.586	0.540	NS	-	52	32	26	-
Le	NS	0.690	0.575	0.498	NS	-	45	30	22	-
1983										
a	NS	0.826	NS	NS	NS	-	67	-	-	-
La	NS	0.706	NS	NS	NS	-	48	-	-	-
c	NS	0.843	NS	NS	NS	-	70	-	-	-
Lc	NS	0.829	NS	NS	NS	-	67	-	-	-
e	NS	0.863	NS	NS	NS	-	73	-	-	-
Le	NS	0.815	NS	NS	NS	-	65	-	-	-

Table J6. Correlations for relation between crop metal and soil metal; potato on calcareous loam.

	correlation coefficient r					percent variance V%				
	Cd	Ni	Cu	Zn	Pb	Cd	Ni	Cu	Zn	Pb
1979										
a	0.570	0.412	0.747	0.713	NS	30	13	54	49	-
La	0.602	0.396	0.755	0.649	NS	34	12	55	40	-
c	0.629	0.485	0.681	0.675	NS	37	20	44	43	-
Lc	0.563	0.470	0.724	0.696	NS	29	19	50	46	-
e	0.615	0.417	0.654	0.623	NS	35	14	40	36	-
Le	0.508	0.415	0.655	0.641	NS	23	14	40	39	-
1980										
a	0.470	NS	0.584	0.414	NS	19	-	31	14	-
La	0.521	NS	0.728	0.576	NS	24	-	51	30	-
c	0.403	NS	0.683	0.540	NS	13	-	44	26	-
Lc	0.455	NS	0.758	0.596	NS	17	-	56	33	-
e	0.415	NS	0.683	NS	NS	14	-	44	-	-
Le	0.448	NS	0.769	NS	NS	17	-	57	-	-
1981										
a	0.608	NS	0.816	0.486	NS	34	-	65	20	-
La	0.652	NS	0.847	0.482	NS	40	-	70	20	-
c	0.618	NS	0.880	0.540	NS	36	-	76	26	-
Lc	0.673	NS	0.921	0.537	NS	43	-	84	26	-
e	0.584	NS	0.854	0.502	NS	31	-	72	22	-
Le	0.676	NS	0.910	0.502	NS	43	-	82	22	-
1982										
a	0.758	NS	0.570	NS	NS	56	-	30	-	-
La	0.782	NS	0.796	NS	NS	59	-	62	-	-
c	0.765	NS	0.586	NS	NS	57	-	32	-	-
Lc	0.661	NS	0.708	NS	NS	41	-	48	-	-
e	0.775	NS	0.585	NS	NS	58	-	32	-	-
Le	0.766	NS	0.772	NS	NS	57	-	58	-	-
1983										
a	NS	0.557	0.443	NS	NS	-	28	16	-	-
La	NS	NS	0.558	NS	NS	-	-	28	-	-
c	NS	0.664	0.472	NS	NS	-	42	19	-	-
Lc	NS	0.539	0.513	NS	NS	-	26	23	-	-
e	NS	0.579	0.474	NS	NS	-	31	19	-	-
Le	0.437	0.400	0.537	NS	NS	16	12	26	-	-

Table J7. Correlations for relation between crop metal and soil metal; lettuce on sandy loam.

	correlation coefficient r					percent variance V%				
	Cd	Ni	Cu	Zn	Pb	Cd	Ni	Cu	Zn	Pb
1979										
a	0.773	NS	0.760	0.829	NS	58	-	56	67	-
La	0.751	NS	0.784	0.793	NS	55	-	60	61	-
c	0.765	0.406	0.733	0.791	NS	57	13	52	61	-
Lc	0.760	0.393	0.808	0.848	NS	56	12	64	71	-
e	0.770	0.573	0.671	0.690	NS	58	30	43	45	-
Le	0.781	0.508	0.778	0.833	NS	59	23	59	68	-
1980										
a	0.688	0.401	0.708	0.728	NS	65	19	60	63	-
La	0.763	NS	0.731	0.825	NS	65	-	52	67	-
c	0.856	NS	0.809	0.819	NS	72	-	64	66	-
Lc	0.794	NS	0.799	0.828	NS	61	-	62	67	-
e	0.822	0.437	0.792	0.807	NS	66	16	61	64	-
Le	0.791	NS	0.787	0.826	NS	61	-	60	67	-
1981										
a	0.859	NS	0.764	0.776	NS	73	-	57	58	-
La	0.807	NS	0.792	0.855	NS	64	-	61	72	-
c	0.851	NS	0.786	0.740	NS	71	-	60	53	-
Lc	0.770	NS	0.818	0.762	NS	58	-	66	56	-
e	0.823	NS	0.746	0.745	NS	66	-	54	54	-
Le	0.762	NS	0.811	0.855	NS	56	-	64	72	-
1982										
a	0.914	0.870	0.695	0.905	NS	83	75	46	81	-
La	0.870	0.743	0.739	0.772	NS	75	53	53	58	-
c	0.893	0.811	0.681	0.887	NS	79	64	44	78	-
Lc	0.795	0.794	0.729	0.883	NS	62	62	51	77	-
e	0.886	0.820	0.686	0.882	NS	78	66	45	77	-
Le	0.794	0.793	0.757	0.844	NS	61	61	56	70	-
1983										
a	0.828	0.733	0.479	0.840	NS	67	52	20	69	-
La	0.790	0.679	0.748	0.827	NS	61	44	54	67	-
c	0.819	0.502	NS	0.771	NS	66	22	-	58	-
Lc	0.724	0.481	0.485	0.784	NS	50	20	20	60	-
e	0.842	0.596	0.532	0.819	NS	70	33	25	66	-
Le	0.814	0.620	0.678	0.905	NS	65	36	44	81	-

Table J8. Correlations for relation between crop metal and soil metal; lettuce on clay.

	correlation coefficient r					percent variance V%				
	Cd	Ni	Cu	Zn	Pb	Cd	Ni	Cu	Zn	Pb
1979										
a	0.709	0.752	NS	0.704	NS	48	55	-	47	-
La	0.621	0.402	NS	0.526	NS	36	13	-	25	-
c	0.768	0.806	NS	0.695	NS	57	63	-	46	-
Lc	0.648	0.772	NS	0.667	NS	40	58	-	42	-
e	0.721	0.824	NS	0.727	NS	50	67	-	51	-
Le	0.622	0.683	NS	0.569	NS	36	44	-	30	-
1980										
a	0.871	0.927	NS	0.910	NS	75	85	-	82	-
La	0.733	0.694	NS	0.686	NS	52	46	-	45	-
c	0.787	0.791	NS	0.812	NS	60	61	-	64	-
Lc	0.658	0.793	NS	0.820	NS	41	61	-	66	-
e	0.791	0.828	NS	0.823	NS	61	67	-	66	-
Le	0.650	0.798	NS	0.751	NS	40	62	-	55	-
1981										
a	0.472	0.918	NS	0.823	NS	19	84	-	66	-
La	NS	0.690	NS	0.827	NS	-	45	-	67	-
c	0.402	0.873	NS	0.795	NS	13	75	-	62	-
Lc	NS	0.855	NS	0.841	NS	-	72	-	69	-
e	0.403	0.873	NS	0.784	NS	13	75	-	60	-
Le	NS	0.769	NS	0.863	NS	-	57	-	73	-
1982										
a	0.759	0.855	NS	0.870	NS	56	72	-	75	-
La	0.601	0.661	NS	0.733	NS	33	41	-	52	-
c	0.678	0.566	NS	0.791	NS	44	29	-	61	-
Lc	0.510	0.552	NS	0.810	NS	23	28	-	64	-
e	0.730	0.911	NS	0.856	NS	51	82	-	72	-
Le	0.558	0.870	NS	0.775	NS	28	75	-	58	-
1983										
a	0.878	0.816	0.410	0.826	0.482	75	65	13	67	20
La	0.725	0.591	NS	0.573	NS	53	35	-	33	-
c	0.835	0.746	NS	0.746	0.499	68	54	-	54	22
Lc	0.708	0.729	NS	0.732	0.467	48	51	-	52	18
e	0.858	0.826	NS	0.806	0.518	72	67	-	63	24
Le	0.724	0.769	NS	0.710	0.467	50	57	-	48	18

Table J9. Correlations for relation between crop metal and soil metal; lettuce on calcareous loam.

	correlation coefficient r					percent variance V%				
	Cd	Ni	Cu	Zn	Pb	Cd	Ni	Cu	Zn	Pb
1979										
a	0.610	NS	NS	0.580	NS	35	-	-	31	-
La	0.707	NS	NS	0.729	NS	48	-	-	51	-
c	0.610	NS	NS	0.631	NS	35	-	-	37	-
Lc	0.727	NS	NS	0.748	NS	51	-	-	54	-
e	0.628	NS	NS	0.604	NS	37	-	-	34	-
Le	0.752	NS	NS	0.747	NS	55	-	-	54	-
1980										
a	0.893	0.776	0.779	0.810	NS	79	59	59	64	-
La	0.915	0.609	0.784	0.697	NS	83	34	60	46	-
c	0.846	0.803	0.759	0.726	NS	70	63	56	51	-
Lc	0.876	0.791	0.865	0.767	NS	76	61	74	57	-
e	0.825	0.766	0.731	0.722	NS	67	57	51	50	-
Le	0.913	0.705	0.822	0.769	NS	83	48	66	57	-
1981										
a	0.815	0.399	NS	NS	NS	65	12	-	-	-
La	0.786	NS	NS	NS	NS	60	-	-	-	-
c	0.763	0.413	NS	NS	NS	56	14	-	-	-
Lc	0.761	NS	NS	NS	NS	56	-	-	-	-
e	0.773	0.410	NS	NS	NS	58	13	-	-	-
Le	0.741	NS	NS	NS	NS	53	-	-	-	-
1982										
a	0.863	NS	NS	NS	NS	73	-	-	-	-
La	0.856	NS	NS	NS	NS	72	-	-	-	-
c	0.908	NS	NS	0.511	NS	82	-	-	23	-
Lc	0.861	NS	NS	0.464	NS	73	-	-	18	-
e	0.884	NS	NS	NS	NS	77	-	-	-	-
Le	0.848	NS	NS	NS	NS	71	-	-	-	-
1983										
a	0.479	NS	NS	NS	NS	20	-	-	-	-
La	0.560	NS	NS	NS	NS	28	-	-	-	-
c	0.596	NS	NS	NS	NS	33	-	-	-	-
Lc	0.577	NS	NS	NS	NS	30	-	-	-	-
e	0.480	NS	NS	NS	NS	26	-	-	-	-
Le	0.487	NS	NS	NS	NS	20	-	-	-	-

Table J10. Correlations for relation between crop metal and soil metal; red beet on sandy loam.

	correlation coefficient r					percent variance V%				
	Cd	Ni	Cu	Zn	Pb	Cd	Ni	Cu	Zn	Pb
1979										
a	0.808	NS	0.438	0.558	NS	64	-	16	28	-
La	0.701	NS	0.447	0.697	NS	47	-	17	46	-
c	0.745	NS	NS	0.447	NS	54	-	-	17	-
Lc	0.622	NS	NS	0.459	NS	36	-	-	18	-
e	0.718	NS	NS	0.408	NS	50	-	-	13	-
Le	0.613	NS	NS	0.580	NS	35	-	-	31	-
1980										
a	0.765	0.666	0.709	0.744	NS	57	42	48	54	-
La	0.858	0.687	0.808	0.797	NS	73	45	64	62	-
c	0.864	0.726	0.699	0.811	NS	74	51	47	64	-
Lc	0.850	0.738	0.769	0.829	NS	71	53	57	67	-
e	0.872	0.740	0.710	0.775	NS	75	53	48	58	-
Le	0.845	0.726	0.772	0.806	NS	70	51	58	64	-
1981										
a	0.935	NS	0.549	0.923	NS	87	-	27	85	-
La	0.911	NS	0.533	0.836	NS	82	-	25	69	-
c	0.952	NS	0.605	0.926	NS	90	-	34	85	-
Lc	0.919	NS	0.639	0.946	NS	84	-	38	89	-
e	0.931	NS	0.626	0.910	NS	86	-	37	82	-
Le	0.914	NS	0.663	0.936	NS	83	-	42	87	-
1982										
a	0.785	0.641	NS	0.713	NS	60	39	-	49	-
La	0.707	0.520	NS	0.644	NS	48	24	-	39	-
c	0.749	0.455	NS	0.659	NS	54	17	-	41	-
Lc	0.696	0.457	NS	0.696	NS	46	18	-	46	-
e	0.771	0.611	NS	0.668	NS	58	35	-	42	-
Le	0.699	0.628	NS	0.707	NS	47	37	-	48	-
1983										
a	0.499	NS	NS	0.721	NS	22	-	-	50	-
La	0.584	NS	NS	0.774	NS	31	-	-	58	-
c	0.401	NS	NS	0.622	NS	13	-	-	36	-
Lc	0.482	NS	NS	0.683	NS	20	-	-	44	-
e	0.410	NS	NS	0.642	NS	13	-	-	39	-
Le	0.540	NS	NS	0.772	NS	26	-	-	58	-

Table J11. Correlations for relation between crop metal and soil metal; red beet on clay.

	correlation coefficient r					percent variance V%				
	Cd	Ni	Cu	Zn	Pb	Cd	Ni	Cu	Zn	Pb
1979										
a	0.767	0.771	NS	0.725	NS	57	58	-	51	-
La	0.714	0.697	NS	0.811	NS	49	46	-	64	-
c	0.725	0.800	NS	0.725	NS	51	62	-	51	-
Lc	0.670	0.824	NS	0.808	NS	43	67	-	64	-
e	0.730	0.813	NS	0.780	NS	51	65	-	59	-
Le	0.657	0.824	NS	0.863	NS	41	67	-	73	-
1980										
a	0.685	0.682	NS	0.599	NS	45	44	-	33	-
La	0.659	0.594	NS	0.747	NS	41	33	-	54	-
c	0.682	0.540	NS	0.566	NS	44	26	-	29	-
Lc	0.577	0.517	NS	0.606	NS	30	24	-	34	-
e	0.692	0.768	NS	0.670	NS	46	57	-	43	-
Le	0.613	0.743	NS	0.756	NS	35	53	-	55	-
1981										
a	0.517	0.764	NS	0.740	NS	24	57	-	53	-
La	0.491	0.570	NS	0.818	NS	21	30	-	65	-
c	0.451	0.493	NS	0.625	NS	17	21	-	37	-
Lc	0.439	0.488	NS	0.680	NS	16	21	-	44	-
e	0.477	0.796	NS	0.640	NS	19	62	-	39	-
Le	0.467	0.724	NS	0.771	NS	19	50	-	58	-
1982										
a	0.660	0.640	NS	0.865	NS	41	38	-	74	-
La	0.565	0.527	NS	0.817	NS	29	25	-	65	-
c	0.630	NS	NS	0.770	NS	37	-	-	58	-
Lc	0.550	NS	NS	0.806	NS	27	-	-	63	-
e	0.641	0.610	NS	0.789	NS	39	35	-	61	-
Le	0.577	0.629	NS	0.849	NS	30	37	-	71	-
1983										
a	0.781	0.946	NS	0.930	NS	59	89	-	86	-
La	0.758	0.773	NS	0.763	NS	56	58	-	57	-
c	0.746	0.853	NS	0.909	NS	54	72	-	82	-
Lc	0.748	0.856	NS	0.926	NS	54	72	-	85	-
e	0.739	0.909	NS	0.907	NS	53	82	-	82	-
Le	0.737	0.906	NS	0.893	NS	52	81	-	79	-

Table J12. Correlations for relation between crop metal and soil metal; red beet on calcareous loam.

	correlation coefficient r					percent variance V%				
	Cd	Ni	Cu	Zn	Pb	Cd	Ni	Cu	Zn	Pb
1979										
a	0.868	0.493	NS	0.448	NS	74	21	-	17	-
La	0.810	NS	NS	0.566	NS	64	-	-	29	-
c	0.701	0.399	NS	NS	NS	47	12	-	-	-
Lc	0.576	NS	NS	NS	NS	30	-	-	-	-
e	0.663	0.519	NS	0.493	NS	41	24	-	21	-
Le	0.528	NS	NS	0.585	NS	25	-	-	31	-
1980										
a	0.823	NS	NS	0.630	NS	66	-	-	37	-
La	0.835	NS	NS	0.432	NS	68	-	-	15	-
c	0.602	NS	NS	0.716	NS	34	-	-	49	-
Lc	0.809	NS	0.390	0.738	NS	64	-	12	53	-
e	0.746	NS	NS	0.661	NS	54	-	-	41	-
Le	0.827	NS	0.405	0.736	NS	67	-	13	52	-
1981										
a	0.858	NS	NS	0.581	NS	70	-	-	31	-
La	0.904	NS	0.565	0.783	NS	78	-	29	60	-
c	0.879	NS	0.390	0.631	NS	76	-	12	37	-
Lc	0.899	NS	0.501	0.725	NS	80	-	22	51	-
e	0.876	NS	NS	0.641	NS	76	-	-	39	-
Le	0.928	NS	0.518	0.765	NS	86	-	24	57	-
1982										
a	0.878	NS	NS	0.621	NS	76	-	-	36	-
La	0.808	NS	NS	0.551	NS	64	-	-	27	-
c	0.862	NS	0.394	0.673	NS	73	-	12	43	-
Lc	0.812	NS	0.467	0.721	NS	65	-	18	50	-
e	0.877	NS	0.416	0.735	NS	76	-	14	52	-
Le	0.779	NS	0.470	0.693	NS	59	-	19	46	-
1983										
a	0.764	NS	NS	0.648	NS	57	-	-	40	-
La	0.774	NS	0.609	0.703	NS	58	-	34	47	-
c	0.773	NS	NS	0.718	NS	58	-	-	49	-
Lc	0.771	NS	0.536	0.792	NS	58	-	26	61	-
e	0.800	NS	0.439	0.735	NS	62	-	16	52	-
Le	0.828	NS	0.617	0.824	NS	67	-	36	67	-

Table J13. Correlations for relation between crop metal and soil metal; cabbage on sandy loam.

	correlation coefficient r					percent variance V%				
	Cd	Ni	Cu	Zn	Pb	Cd	Ni	Cu	Zn	Pb
1979										
a	NS	NS	0.738	0.747	0.432	-	-	53	54	15
La	NS	NS	0.853	0.678	0.453	-	-	72	44	17
c	NS	NS	0.727	0.677	0.553	-	-	51	44	28
Lc	NS	NS	0.814	0.694	0.531	-	-	65	46	25
e	NS	NS	0.680	0.668	0.444	-	-	44	42	16
Le	NS	NS	0.809	0.714	0.481	-	-	64	49	20
1980										
a	0.698	NS	0.709	0.715	NS	47	-	48	49	-
La	0.649	NS	0.717	0.841	NS	40	-	49	69	-
c	0.714	NS	0.640	0.717	NS	49	-	39	49	-
Lc	0.646	NS	0.707	0.790	NS	39	-	48	61	-
e	0.665	NS	0.624	0.784	NS	42	-	36	60	-
Le	0.632	NS	0.695	0.850	NS	37	-	46	71	-
1981										
a	0.894	0.812	0.724	0.791	NS	79	65	50	61	-
La	0.780	0.857	0.680	0.848	NS	59	72	44	71	-
c	0.872	0.713	0.606	0.697	NS	75	49	34	46	-
Lc	0.686	0.716	0.669	0.765	NS	45	49	42	57	-
e	0.866	0.733	0.573	0.667	NS	74	52	30	42	-
Le	0.712	0.805	0.644	0.817	NS	49	63	39	65	-
1982										
a	0.511	0.886	0.695	0.900	NS	23	78	46	80	-
La	0.513	0.781	0.520	0.834	NS	23	59	24	68	-
c	0.400	0.725	0.729	0.868	NS	12	51	51	74	-
Lc	0.447	0.712	0.672	0.899	NS	17	49	43	80	-
e	NS	0.802	0.705	0.863	NS	-	63	48	73	-
Le	0.452	0.821	0.626	0.903	NS	17	66	37	81	-
1983										
a	0.629	NS	NS	0.448	NS	44	-	-	16	-
La	0.639	NS	NS	NS	NS	40	-	-	-	-
c	0.679	NS	NS	0.398	NS	43	-	-	11	-
Lc	0.642	NS	NS	0.416	NS	38	-	-	12	-
e	0.656	NS	NS	NS	NS	39	-	-	-	-
Le	0.600	NS	NS	0.421	NS	32	-	-	13	-

Table J14. Correlations for relation between crop metal and soil metal; cabbage on clay.

	correlation coefficient r					percent variance V%				
	Cd	Ni	Cu	Zn	Pb	Cd	Ni	Cu	Zn	Pb
1979										
a	NS	0.932	0.794	0.637	NS	-	86	62	38	-
La	NS	0.732	0.854	0.625	NS	-	52	72	37	-
c	NS	0.870	0.773	0.577	NS	-	75	58	31	-
Lc	NS	0.891	0.860	0.643	NS	-	79	73	39	-
e	NS	0.849	0.753	0.565	NS	-	71	55	29	-
Le	NS	0.854	0.855	0.652	NS	-	72	72	40	-
1980										
a	0.662	0.914	0.703	0.848	NS	41	83	47	71	-
La	0.640	0.750	0.783	0.924	NS	38	54	60	85	-
c	0.608	0.901	0.689	0.816	NS	34	80	45	65	-
Lc	0.615	0.910	0.768	0.890	NS	35	82	57	78	-
e	0.637	0.907	0.701	0.807	NS	38	81	47	64	-
Le	0.627	0.861	0.784	0.948	NS	37	73	60	90	-
1981										
a	0.578	0.764	0.827	0.834	NS	31	57	67	68	-
La	0.496	0.566	0.792	0.798	NS	21	29	61	62	-
c	0.586	0.505	0.675	0.706	NS	32	22	43	48	-
Lc	0.480	0.482	0.744	0.733	NS	20	20	54	52	-
e	0.615	0.786	0.718	0.792	NS	35	60	50	61	-
Le	0.482	0.742	0.780	0.814	NS	20	53	59	65	-
1982										
a	0.513	0.866	0.592	0.880	NS	23	74	32	77	-
La	NS	0.612	0.593	0.695	NS	-	35	32	46	-
c	0.490	0.795	0.513	0.826	NS	21	62	23	67	-
Lc	NS	0.749	0.546	0.820	NS	-	54	27	66	-
e	0.442	0.890	0.544	0.840	NS	16	78	27	69	-
Le	NS	0.789	0.572	0.781	NS	-	61	30	59	-
1983										
a	0.393	0.910	0.768	0.736	NS	12	82	57	52	-
La	NS	0.724	0.572	0.809	NS	-	50	30	64	-
c	NS	0.824	0.815	0.713	NS	-	67	65	49	-
Lc	NS	0.808	0.795	0.766	NS	-	64	62	57	-
e	NS	0.918	0.796	0.712	NS	-	84	62	49	-
Le	NS	0.859	0.715	0.819	NS	-	73	49	66	-

Table J15. Correlations for relation between crop metal and soil metal; cabbage on calcareous loam.

	correlation coefficient r					percent variance V%				
	Cd	Ni	Cu	Zn	Pb	Cd	Ni	Cu	Zn	Pb
1979										
a	0.864	0.838	0.708	0.898	NS	74	69	48	80	-
La	0.850	0.806	0.859	0.785	NS	71	64	73	60	-
c	0.867	0.859	0.665	0.851	NS	74	73	42	71	-
Lc	0.837	0.887	0.783	0.873	NS	69	78	60	75	-
e	0.878	0.858	0.708	0.859	NS	76	73	48	73	-
Le	0.857	0.867	0.766	0.848	NS	72	74	57	71	-
1980										
a	0.924	0.892	0.803	0.913	NS	85	79	63	83	-
La	0.854	0.668	0.807	0.775	NS	72	42	64	58	-
c	0.882	0.930	0.889	0.918	NS	77	86	78	84	-
Lc	0.826	0.885	0.920	0.929	NS	67	77	84	86	-
e	0.887	0.946	0.883	0.927	NS	78	89	77	85	-
Le	0.823	0.746	0.895	0.891	NS	66	54	79	79	-
1981										
a	0.663	0.543	0.593	0.536	NS	40	27	32	26	-
La	0.634	0.488	0.604	0.454	NS	36	21	34	17	-
c	0.559	0.577	0.645	0.773	NS	28	31	39	58	-
Lc	0.551	0.579	0.707	0.769	NS	27	31	48	57	-
e	0.551	0.568	0.707	0.786	NS	27	29	48	60	-
Le	0.550	0.504	0.751	0.768	NS	27	22	55	57	-
1982										
a	NS	0.496	NS	NS	NS	-	21	-	-	-
La	NS	0.543	0.458	NS	NS	-	26	18	-	-
c	NS	0.568	0.464	0.434	NS	-	29	18	15	-
Lc	NS	0.579	0.495	0.466	NS	-	31	21	18	-
e	NS	0.535	0.475	0.422	NS	-	26	19	14	-
Le	NS	0.568	0.576	0.480	NS	-	29	30	20	-
1983										
a	0.471	0.759	0.472	0.439	NS	19	56	19	16	-
La	0.530	0.449	0.583	0.645	NS	25	17	31	39	-
c	0.565	0.452	0.592	0.570	NS	29	17	32	30	-
Lc	0.640	0.404	0.671	0.608	NS	38	13	43	34	-
e	0.610	0.555	0.606	0.641	NS	35	28	34	39	-
Le	0.662	0.458	0.688	0.714	NS	42	18	45	49	-

Table J16. Correlations for relation between crop metal and soil metal; ryegrass on sandy loam.

	correlation coefficient r					percent variance V%				
	Cd	Ni	Cu	Zn	Pb	Cd	Ni	Cu	Zn	Pb
1979										
a	0.890	0.894	0.754	0.794	NS	74	79	55	62	-
La	0.868	0.858	0.874	0.894	NS	72	72	75	79	-
c	0.854	0.853	0.748	0.739	NS	72	72	54	53	-
Lc	0.840	0.853	0.848	0.800	NS	69	72	71	62	-
e	0.849	0.914	0.719	0.760	0.431	71	83	50	56	15
Le	0.828	0.935	0.833	0.885	0.392	67	87	68	78	12
1980										
a	0.854	0.838	0.707	0.829	NS	72	69	48	67	-
La	0.899	0.841	0.816	0.915	NS	80	70	65	83	-
c	0.761	0.802	0.664	0.719	NS	56	63	42	50	-
Lc	0.724	0.810	0.709	0.759	NS	50	64	48	56	-
e	0.758	0.852	0.629	0.657	NS	56	71	37	41	-
Le	0.718	0.883	0.675	0.665	NS	50	77	43	42	-
1981										
a	0.911	0.925	0.838	0.864	NS	82	85	69	74	-
La	0.919	0.873	0.913	0.920	NS	84	75	83	84	-
c	0.871	0.843	0.859	0.876	NS	75	70	73	76	-
Lc	0.846	0.844	0.927	0.919	NS	70	70	85	84	-
e	0.857	0.831	0.866	0.844	NS	72	68	74	70	-
Le	0.848	0.821	0.936	0.954	NS	71	66	87	91	-
1982										
a	0.935	0.891	0.800	0.900	NS	87	78	63	80	-
La	0.935	0.848	0.807	0.902	NS	87	71	64	81	-
c	0.938	0.826	0.859	0.899	NS	87	67	73	80	-
Lc	0.863	0.820	0.872	0.941	NS	73	66	75	88	-
e	0.940	0.870	0.709	0.886	NS	88	75	48	78	-
Le	0.883	0.889	0.698	0.950	NS	77	78	47	90	-
1983										
a	0.970	0.943	0.880	0.936	0.695	94	88	77	87	46
La	0.876	0.808	0.843	0.860	0.720	76	68	70	73	50
c	0.961	0.816	0.928	0.964	0.617	92	65	86	93	36
Lc	0.803	0.797	0.936	0.973	0.666	63	62	87	94	42
e	0.970	0.940	0.906	0.943	0.673	94	88	81	88	43
Le	0.825	0.933	0.915	0.955	0.725	67	86	83	91	51

Table J17. Correlations for relation between crop metal and soil metal; ryegrass on clay.

	correlation coefficient r					percent variance V%				
	Cd	Ni	Cu	Zn	Pb	Cd	Ni	Cu	Zn	Pb
1979										
a	0.788	0.838	0.751	0.793	0.493	61	69	55	61	21
La	0.788	0.753	0.864	0.845	NS	60	55	74	70	-
c	0.796	0.827	0.736	0.794	0.400	62	67	52	62	13
Lc	0.793	0.833	0.830	0.839	NS	61	68	68	69	-
e	0.814	0.833	0.757	0.810	0.417	65	68	55	64	14
Le	0.807	0.839	0.853	0.866	NS	64	69	72	74	-
1980										
a	0.605	0.826	0.596	0.775	NS	34	67	33	58	-
La	0.760	0.752	0.620	0.881	NS	56	55	36	77	-
c	0.529	0.627	0.424	0.616	NS	25	37	15	35	-
Lc	0.649	0.605	0.394	0.647	NS	40	34	12	39	-
e	0.570	0.723	0.439	0.635	NS	30	50	16	38	-
Le	0.697	0.689	0.392	0.671	NS	46	45	12	43	-
1981										
a	0.788	0.863	0.751	0.826	NS	61	73	55	67	-
La	0.896	0.848	0.730	0.895	NS	80	71	51	79	-
c	0.762	0.680	0.798	0.840	NS	56	44	62	69	-
Lc	0.774	0.667	0.816	0.875	NS	58	42	65	75	-
e	0.781	0.801	0.839	0.869	NS	59	63	69	75	-
Le	0.775	0.802	0.828	0.886	NS	58	63	67	78	-
1982										
a	0.865	0.874	0.844	0.839	NS	74	75	70	69	-
La	0.934	0.883	0.791	0.879	NS	87	77	61	76	-
c	0.781	0.746	0.772	0.802	NS	59	54	58	63	-
Lc	0.899	0.759	0.847	0.871	NS	80	56	71	75	-
e	0.835	0.862	0.816	0.863	NS	69	73	65	73	-
Le	0.892	0.912	0.854	0.919	NS	79	83	72	84	-
1983										
a	0.921	0.885	0.830	0.863	NS	84	77	68	73	-
La	0.915	0.841	0.840	0.870	NS	83	70	69	75	-
c	0.875	0.746	0.806	0.816	NS	76	54	63	65	-
Lc	0.881	0.741	0.884	0.877	NS	77	53	77	76	-
e	0.902	0.867	0.831	0.884	NS	81	74	68	77	-
Le	0.871	0.884	0.899	0.922	NS	75	77	80	84	-

Table J18. Correlations for relation between crop metal and soil metal; ryegrass on calcareous loam.

	correlation coefficient r					percent variance V%				
	Cd	Ni	Cu	Zn	Pb	Cd	Ni	Cu	Zn	Pb
1979										
a	0.881	0.922	0.831	0.893	NS	77	84	68	79	-
La	0.900	0.826	0.877	0.881	NS	80	67	76	77	-
c	0.842	0.816	0.835	0.837	NS	70	65	69	69	-
Lc	0.835	0.826	0.912	0.927	NS	68	67	83	85	-
e	0.805	0.781	0.815	0.747	NS	63	59	65	54	-
Le	0.841	0.834	0.897	0.933	NS	69	68	80	87	-
1980										
a	0.863	0.955	0.718	0.885	NS	73	91	50	77	-
La	0.885	0.787	0.705	0.870	NS	77	60	48	75	-
c	0.785	0.898	0.657	0.753	NS	60	80	41	55	-
Lc	0.853	0.906	0.741	0.872	NS	72	81	53	75	-
e	0.796	0.902	0.671	0.763	NS	62	81	43	56	-
Le	0.893	0.894	0.756	0.923	NS	79	79	55	85	-
1981										
a	0.946	0.976	0.809	0.934	NS	89	95	64	87	-
La	0.946	0.804	0.874	0.868	NS	89	63	75	74	-
c	0.923	0.915	0.853	0.902	NS	84	83	72	81	-
Lc	0.916	0.846	0.928	0.935	NS	83	70	86	87	-
e	0.953	0.928	0.874	0.908	NS	90	86	75	82	-
Le	0.943	0.742	0.933	0.949	NS	88	53	87	90	-
1982										
a	0.904	0.947	0.829	0.885	NS	81	89	67	77	-
La	0.829	0.749	0.842	0.854	NS	67	54	70	72	-
c	0.911	0.917	0.883	0.901	NS	82	83	77	80	-
Lc	0.829	0.866	0.920	0.923	NS	67	74	84	85	-
e	0.884	0.952	0.876	0.903	NS	77	90	76	81	-
Le	0.793	0.830	0.910	0.933	NS	61	68	82	87	-
1983										
a	0.798	0.860	0.886	0.858	NS	62	73	78	73	-
La	0.724	0.732	0.841	0.838	NS	50	52	70	69	-
c	0.812	0.842	0.835	0.795	NS	65	70	68	62	-
Lc	0.734	0.813	0.861	0.842	NS	52	65	73	70	-
e	0.814	0.828	0.845	0.844	NS	65	67	70	70	-
Le	0.699	0.767	0.868	0.887	NS	47	57	74	78	-

APPENDIX K

FIGURES ILLUSTRATING RAW DATA FOR CADMIUM AND ZINC IN CROPS IN RELATION TO SOIL METAL

APPENDIX K LIST OF FIGURES

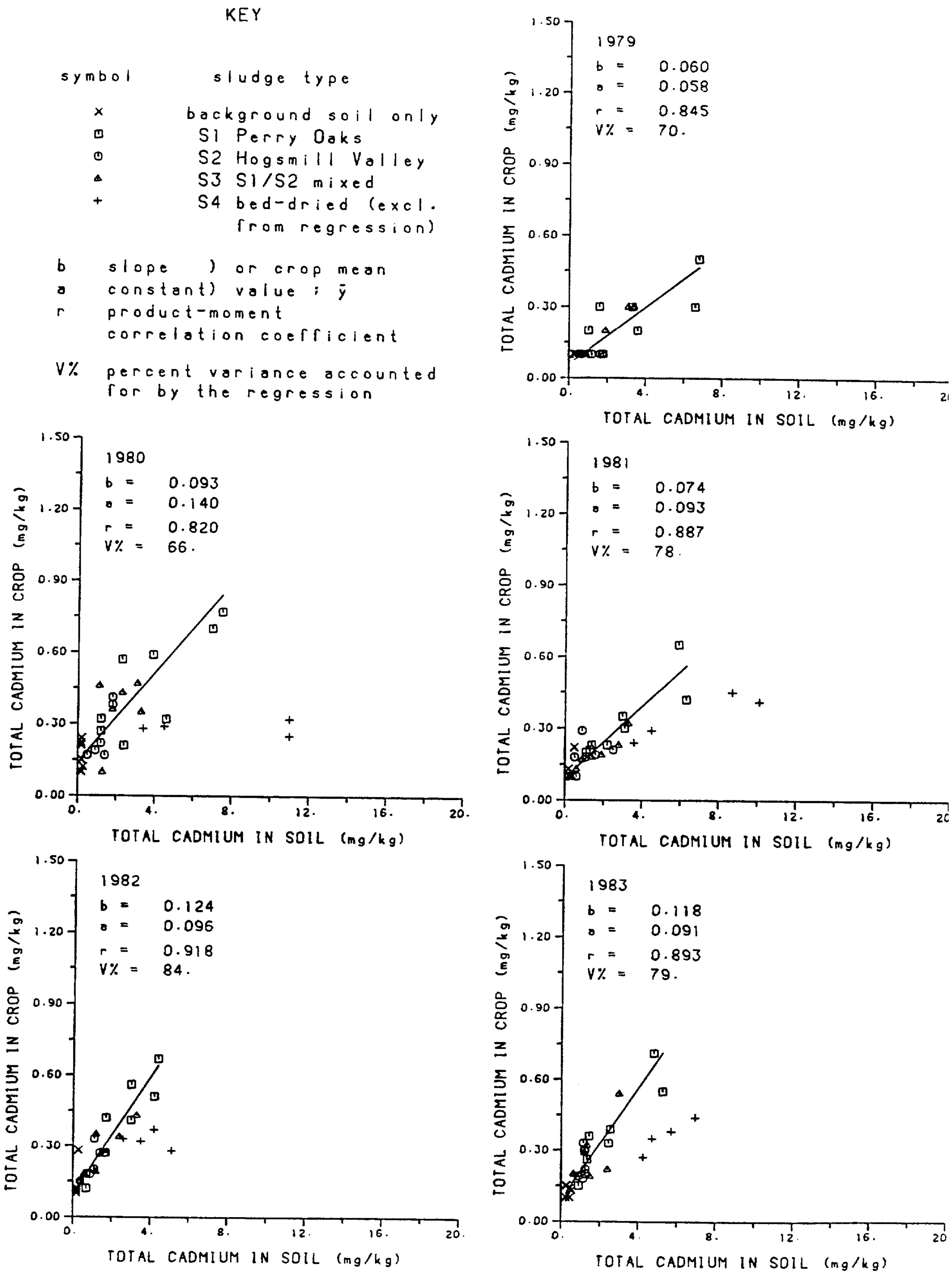


Fig K1. Cadmium in wheat vs. total cadmium in sandy loam 1979-83

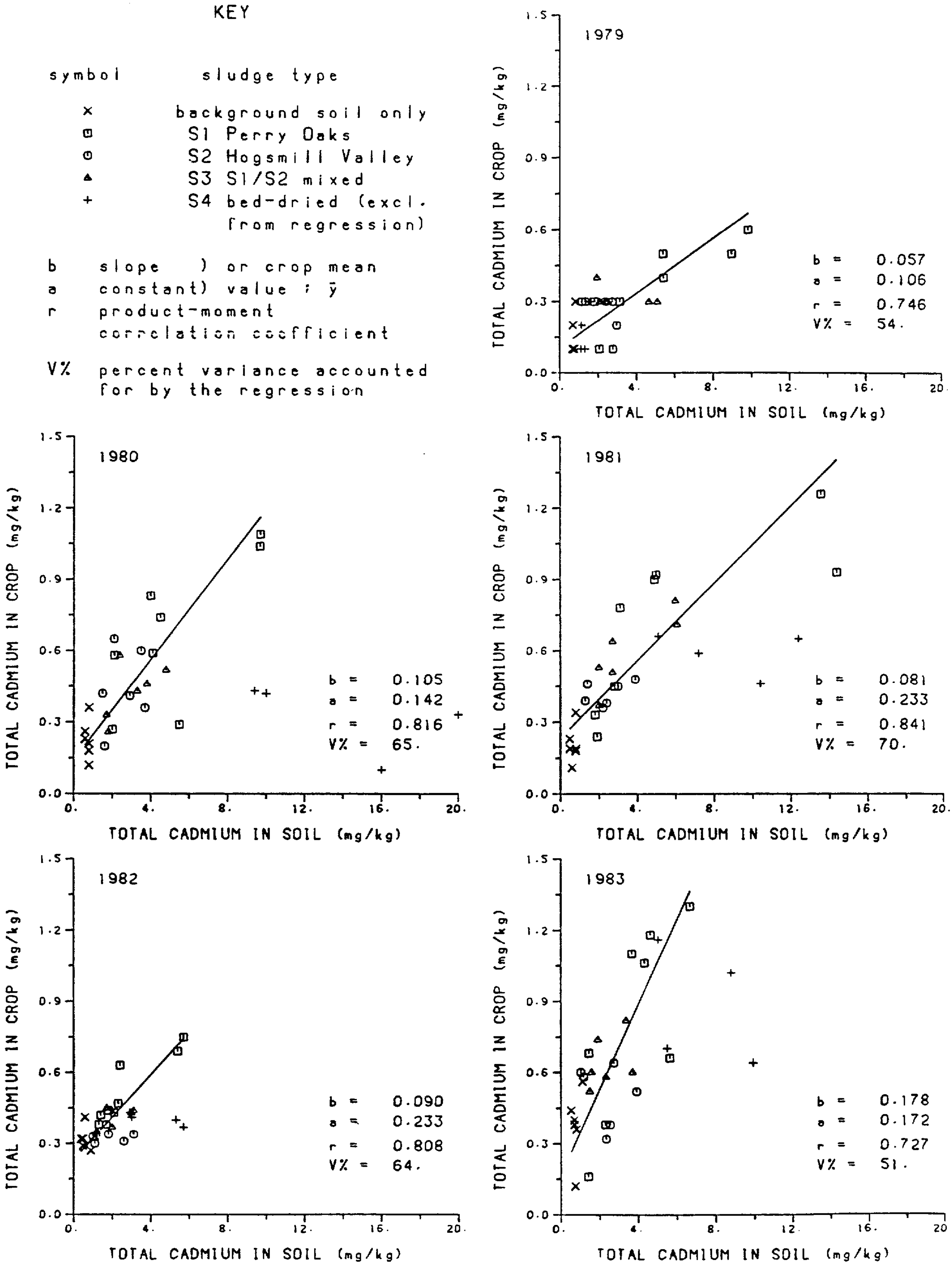


Fig K2. Cadmium in wheat vs. total cadmium in clay 1979-83

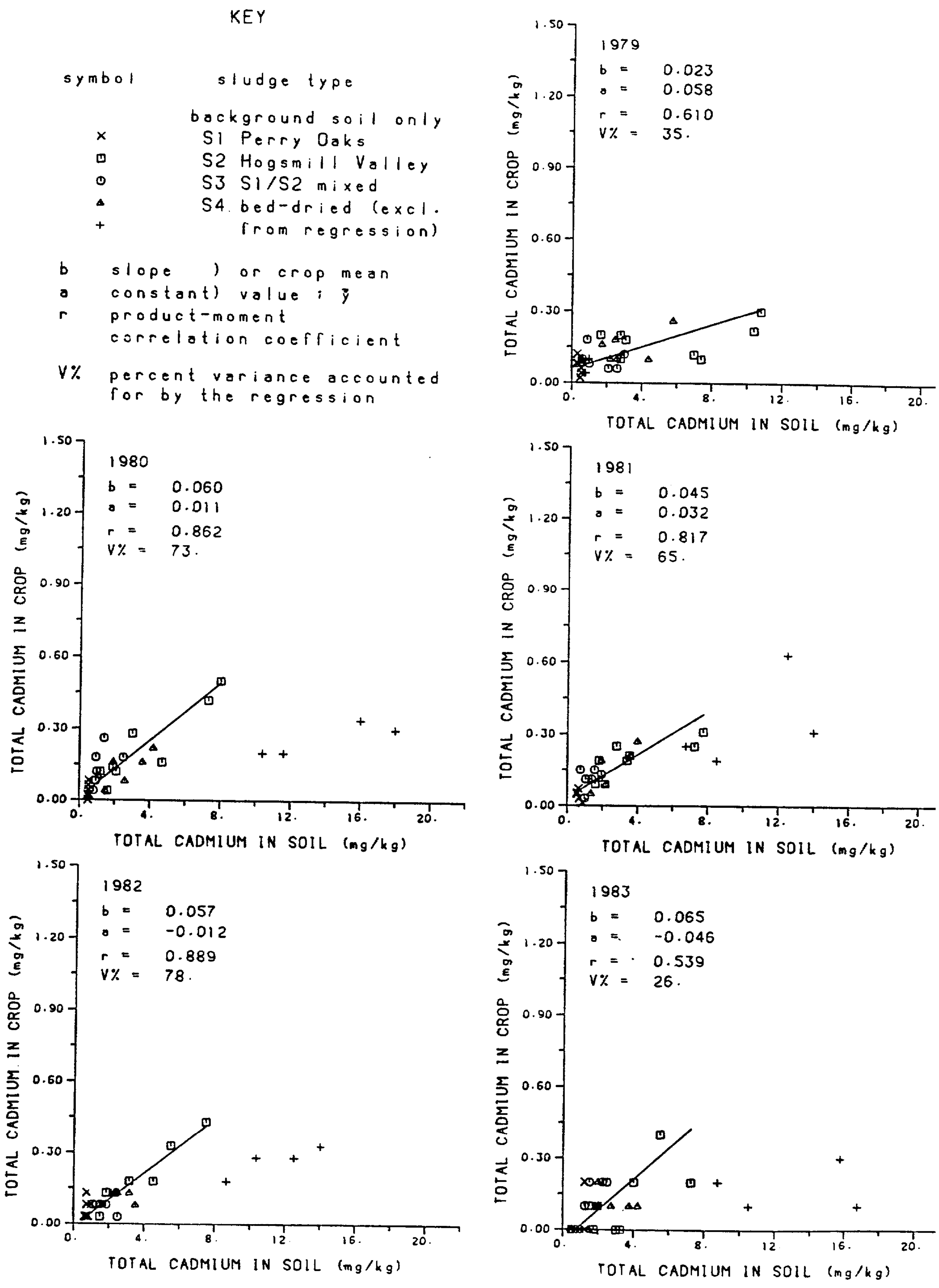


Fig K3. Cadmium in wheat vs. total cadmium in calc. loam 1979-83

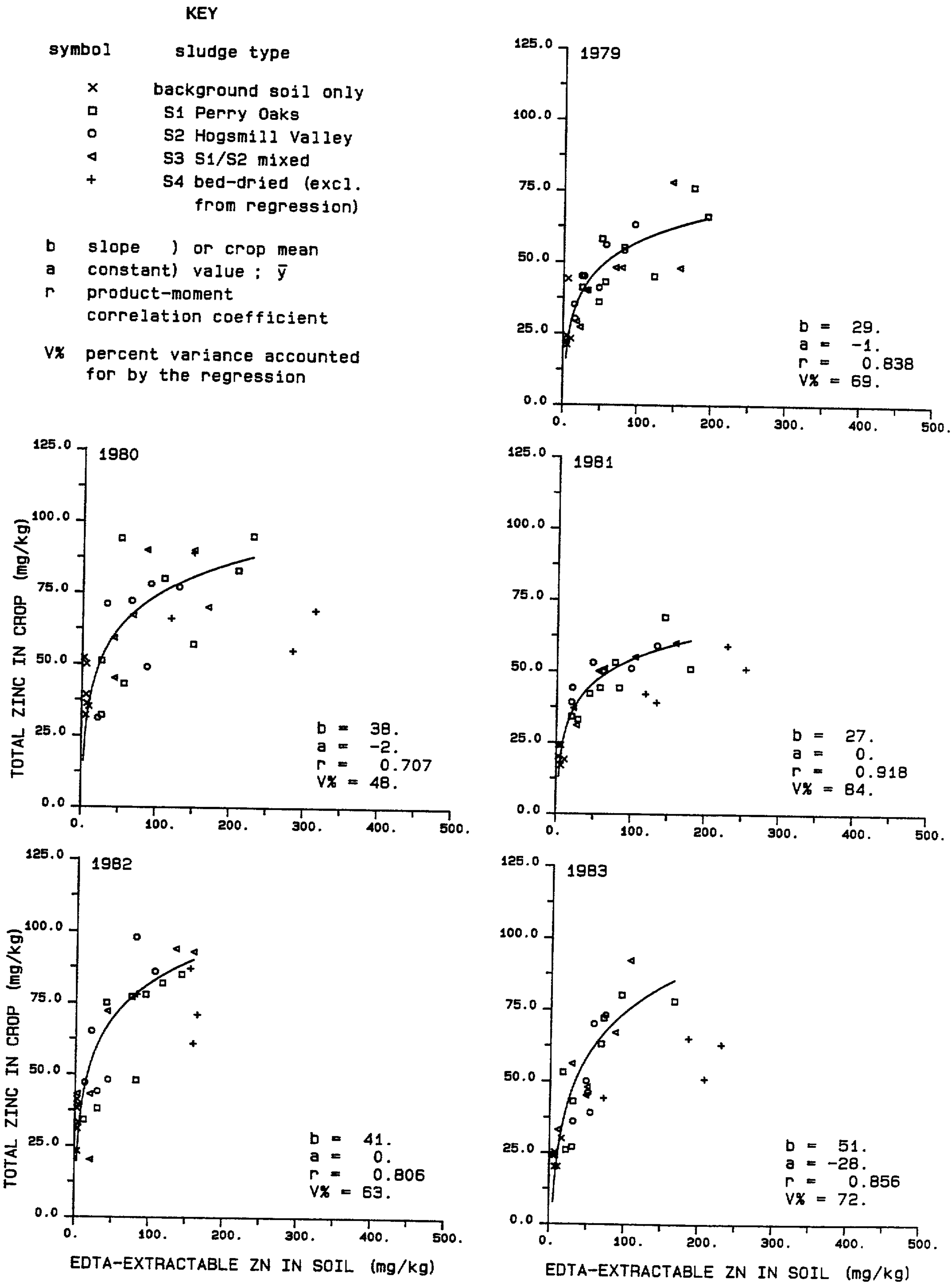


Fig K4. Zinc in wheat vs. EDTA-Zinc in sandy loam 1979-83

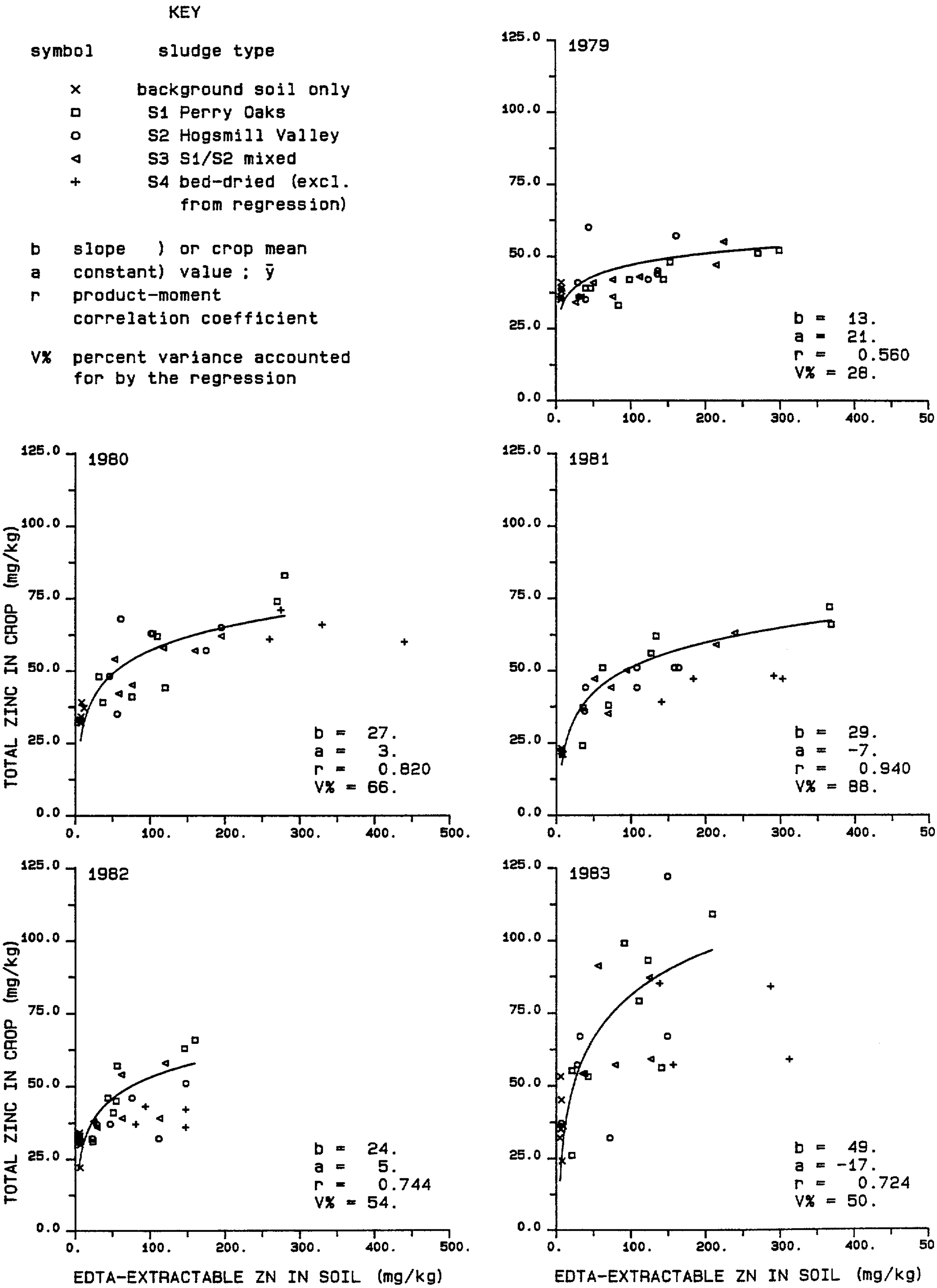


Fig K5. Zinc in wheat vs. EDTA-Zinc in clay 1979-83

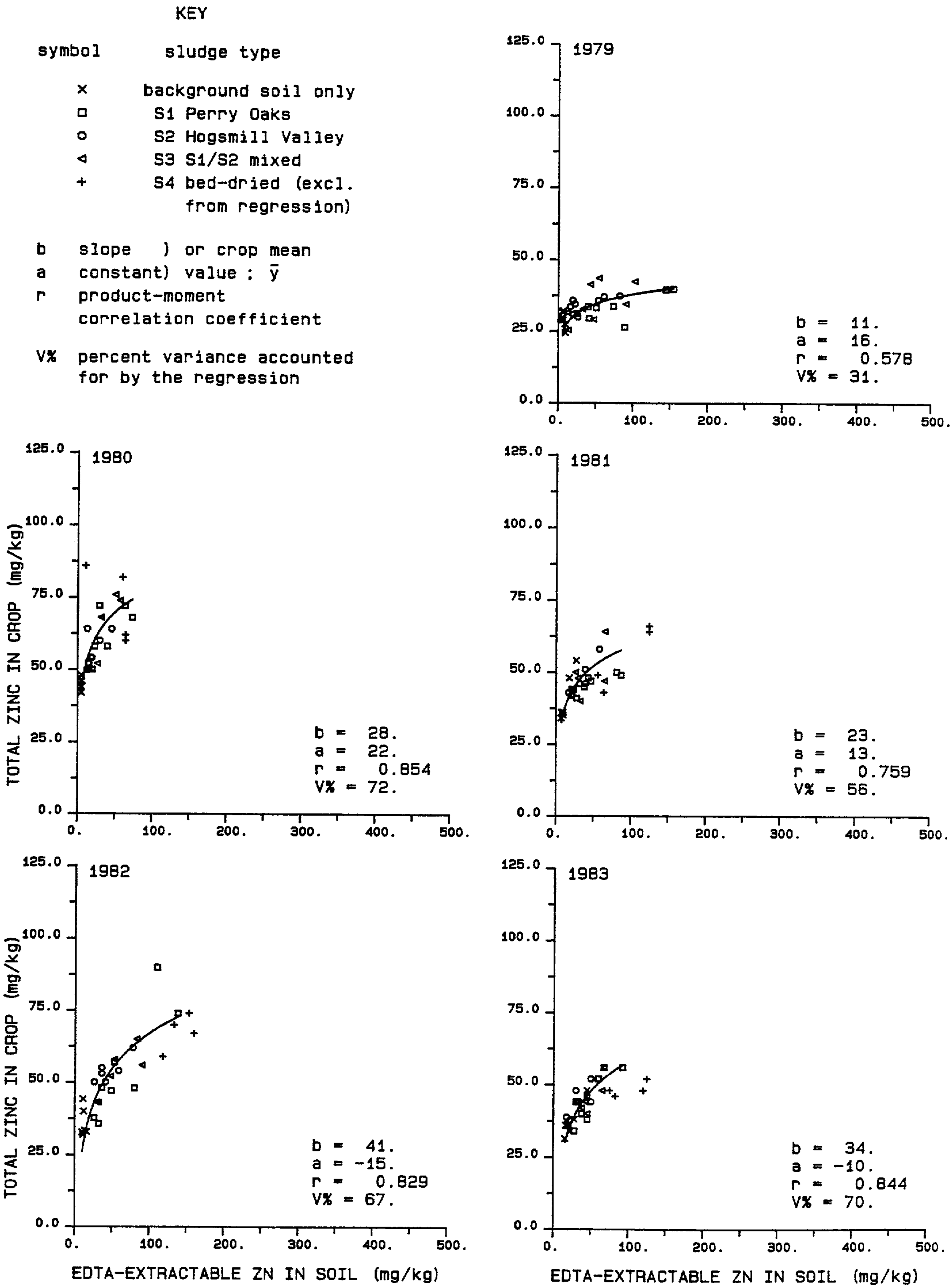


Fig K6. Zinc in wheat vs. EDTA-Zinc in calcareous loam 1979-83

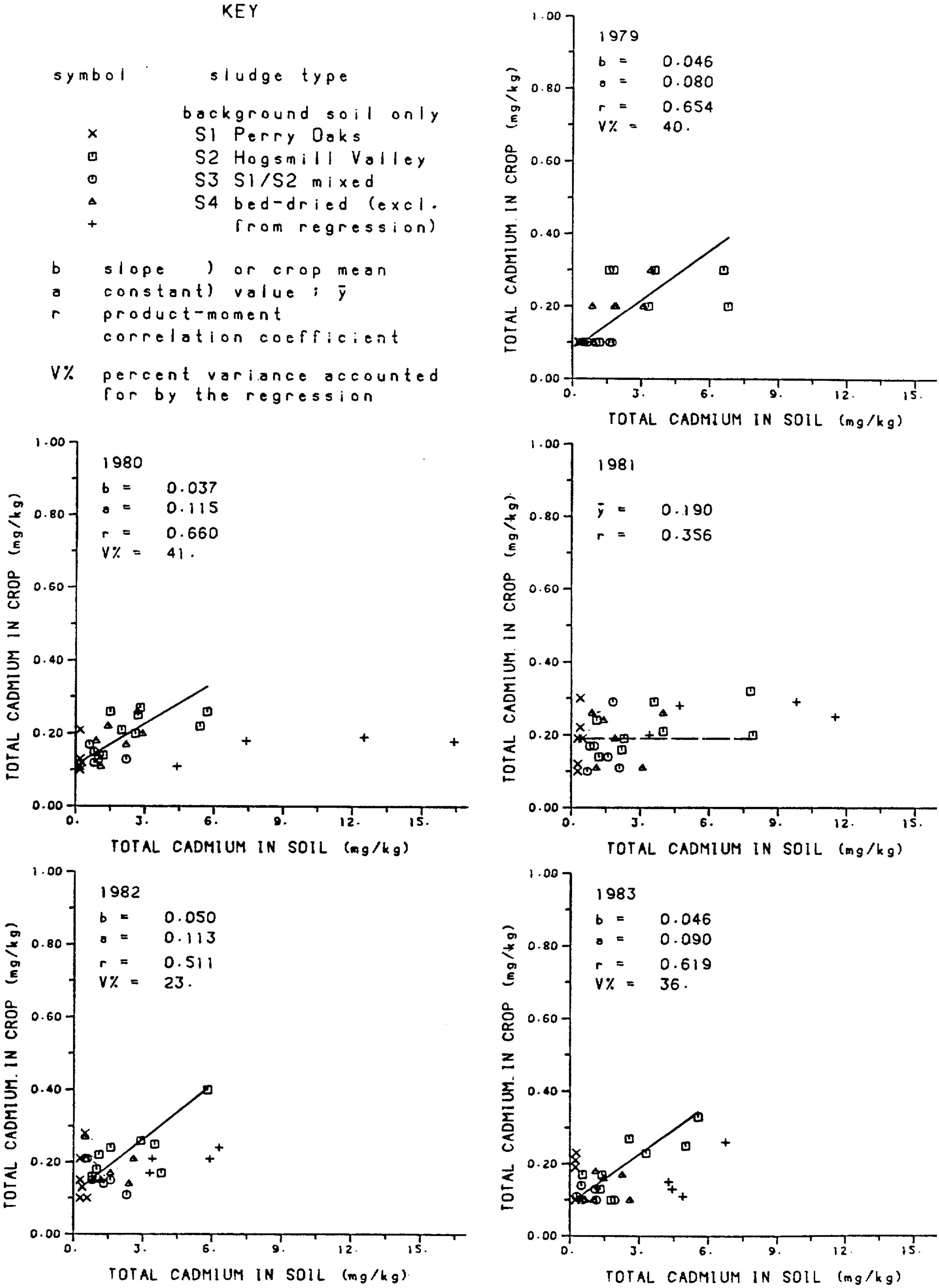


Fig K7. Cadmium in potato vs. total cadmium in sandy loam 1979-83

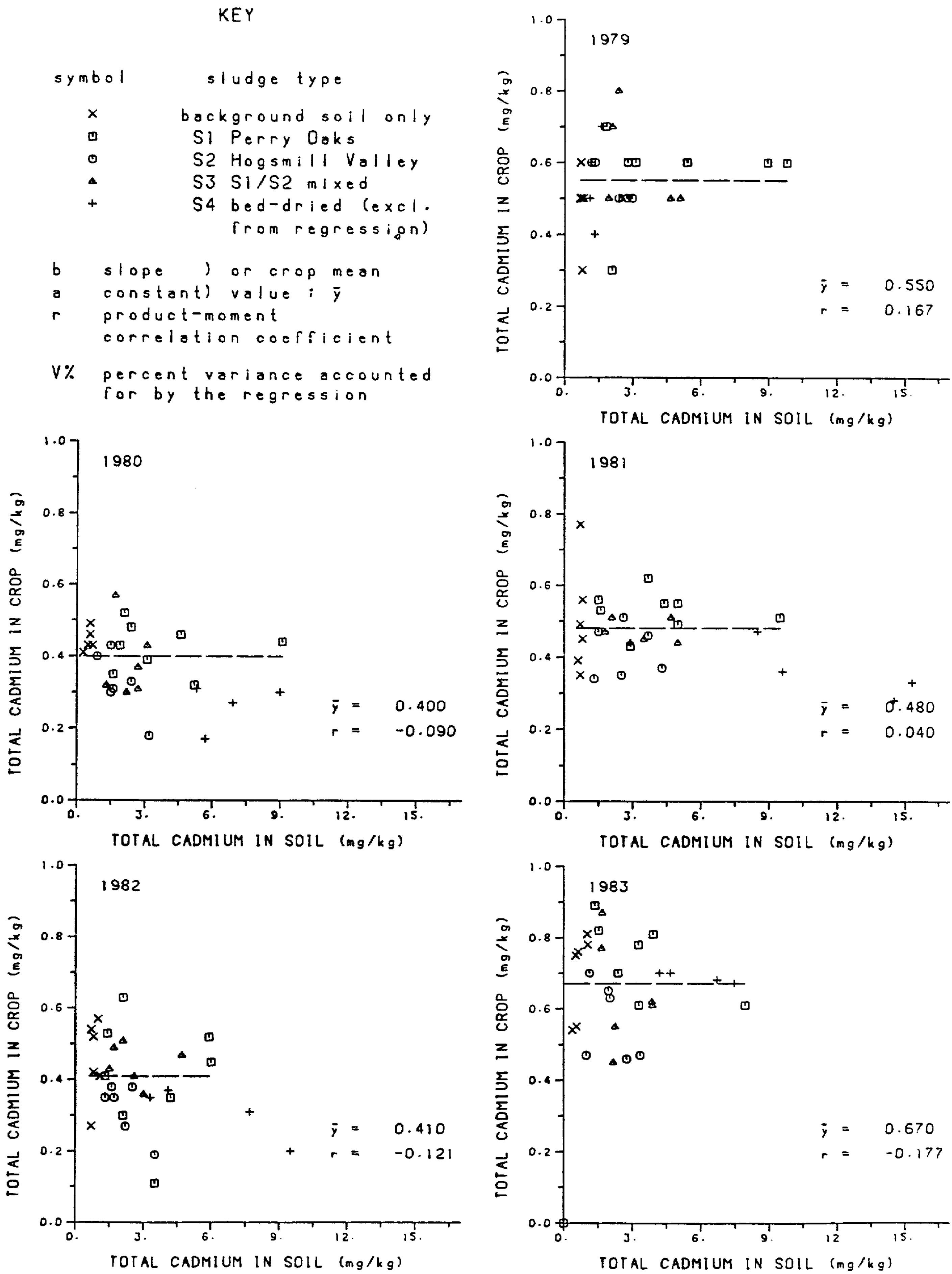


Fig K8. Cadmium in potato vs. total cadmium in clay 1979-83

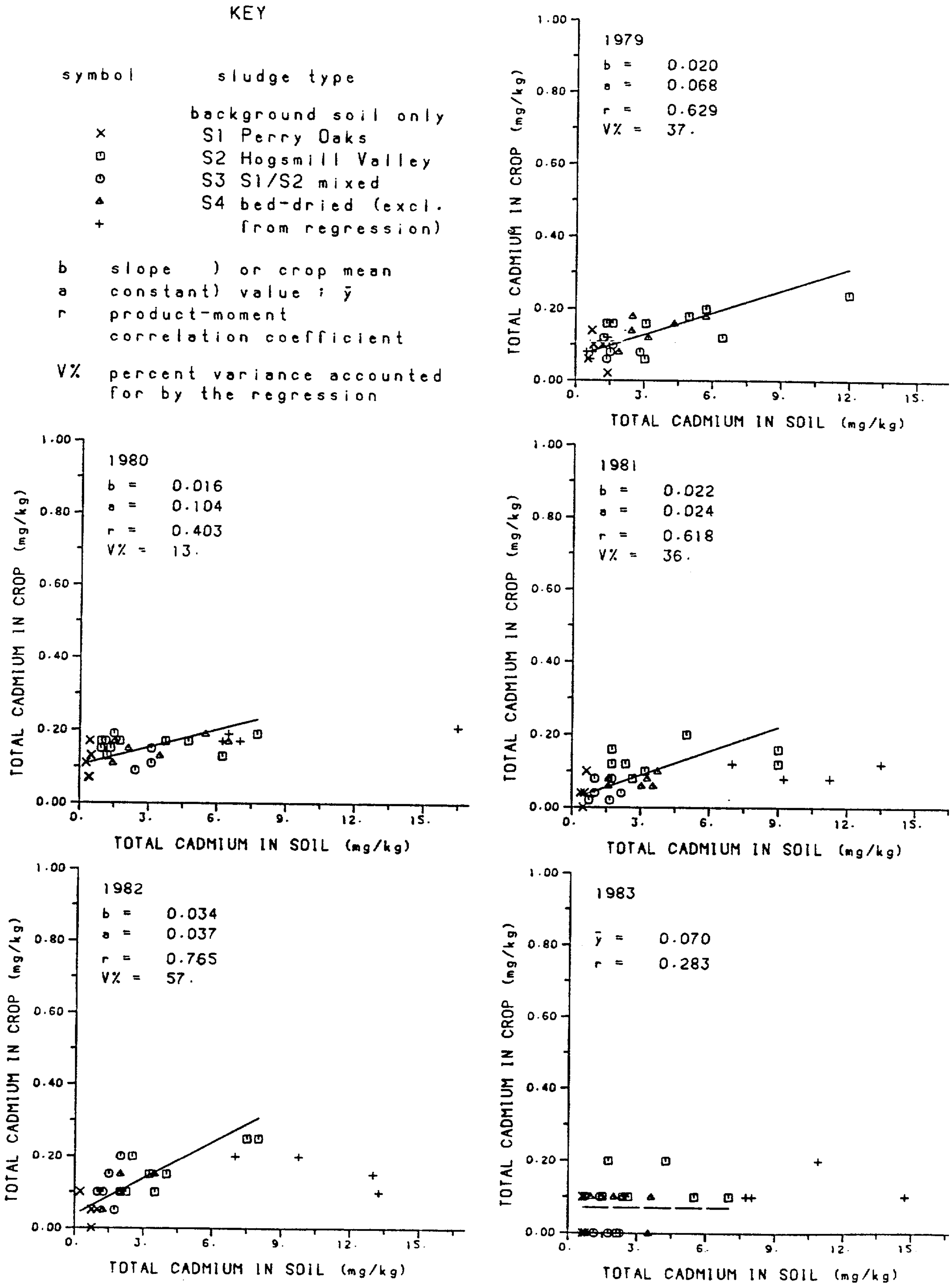


Fig K9. Cadmium in potato vs. total cadmium in calc. loam 1979-83

KEY

symbol	sludge type
×	background soil only
□	S1 Perry Oaks
o	S2 Hogsmill Valley
◁	S3 S1/S2 mixed
+	S4 bed-dried (excl. from regression)

b slope) or crop mean
a constant) value : $\bar{y}$
r product-moment correlation coefficient

V% percent variance accounted for by the regression

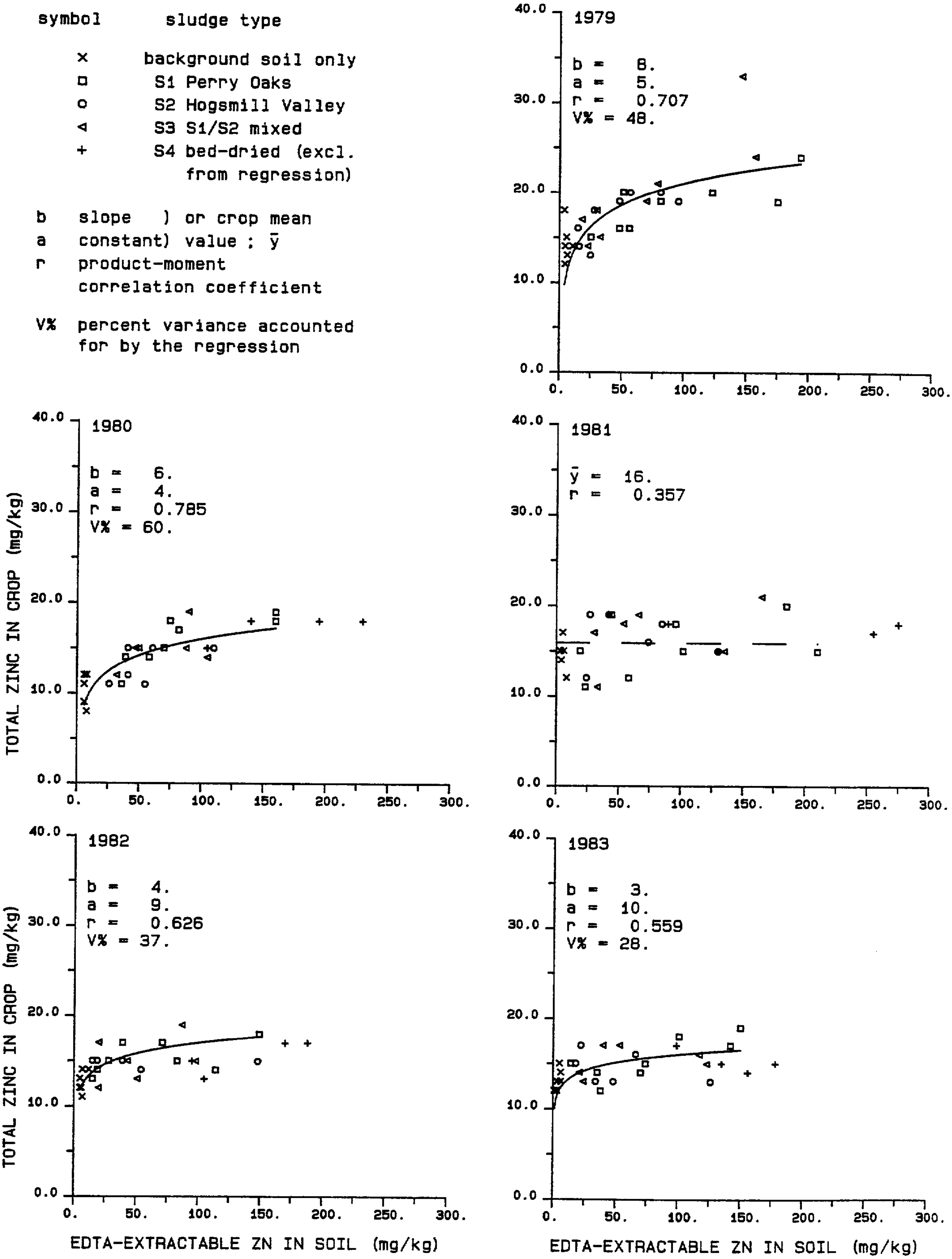


Fig K10. Zinc in potato vs. EDTA-Zinc in sandy loam 1979-83

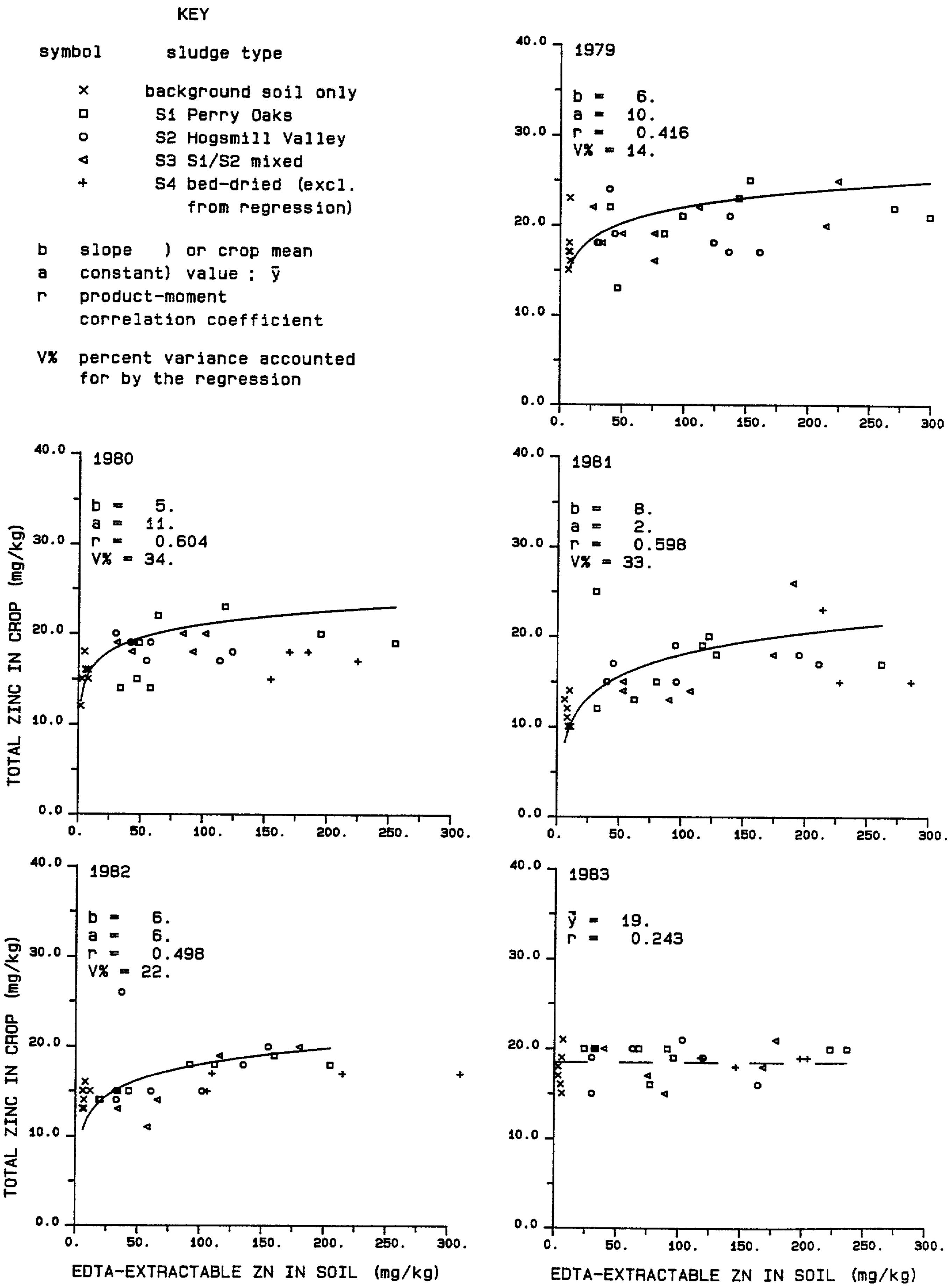


Fig K11. Zinc in potato vs. EDTA-Zinc in clay 1979-83

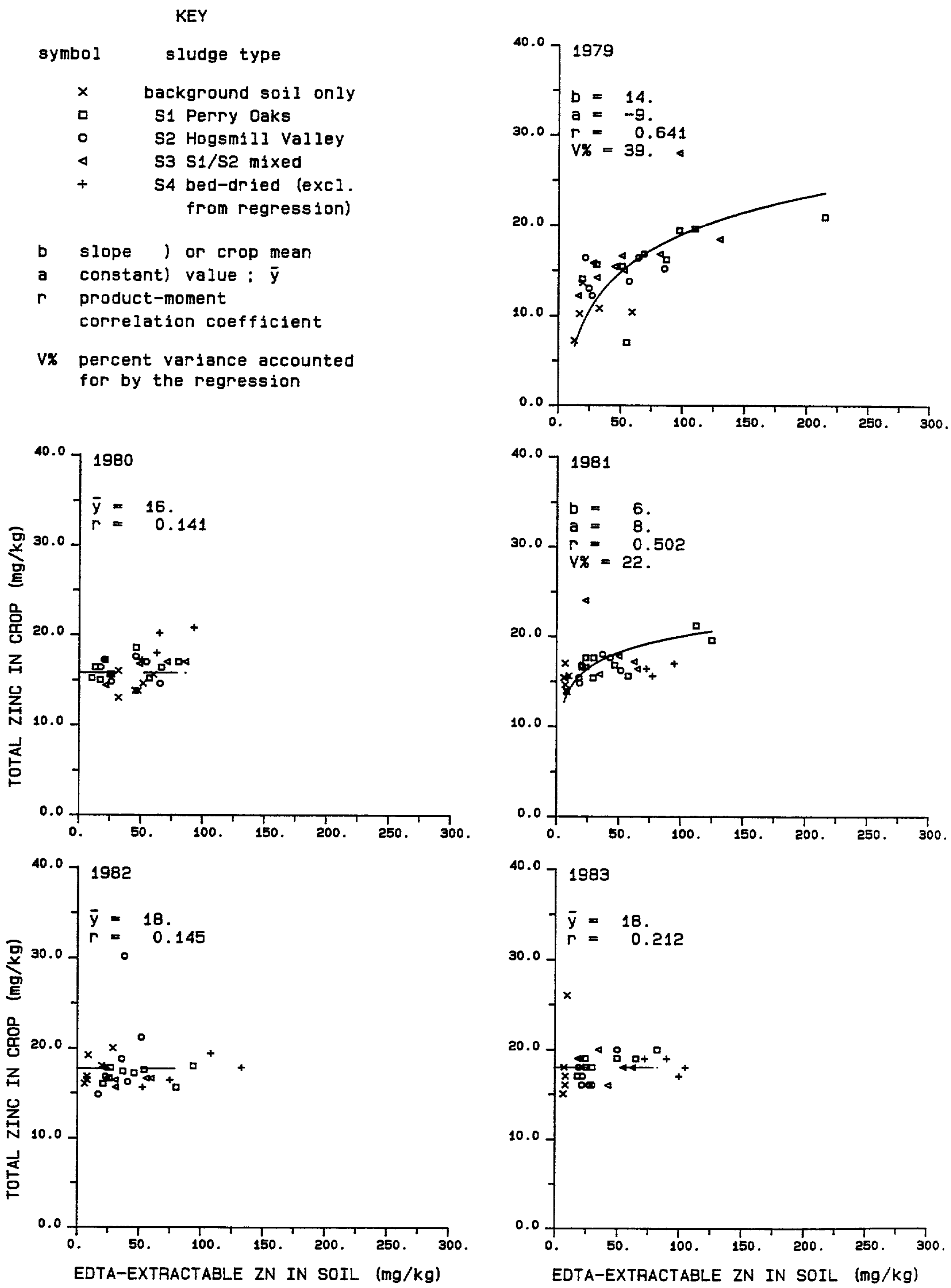
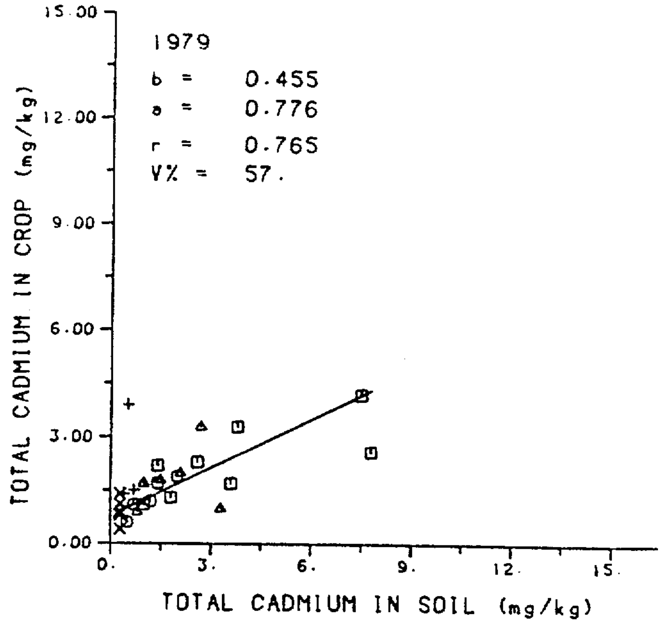


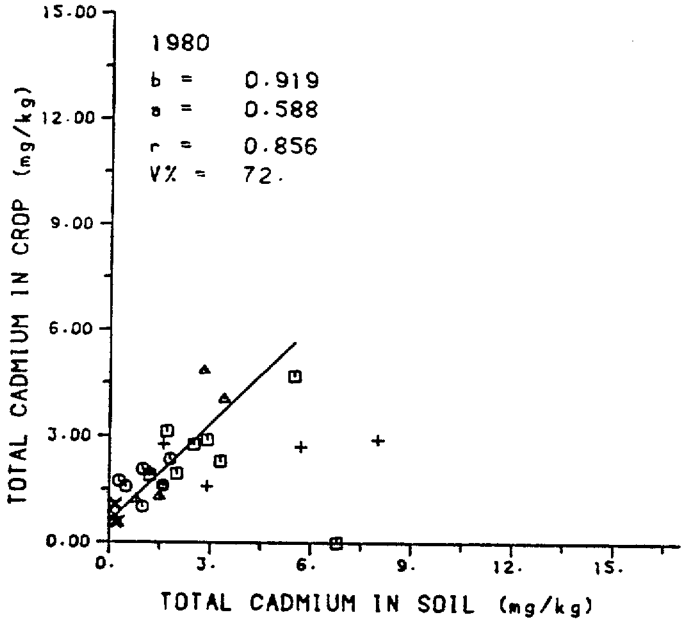
Fig K12. Zinc in potato vs. EDTA-Zinc in calcareous loam 1979-83

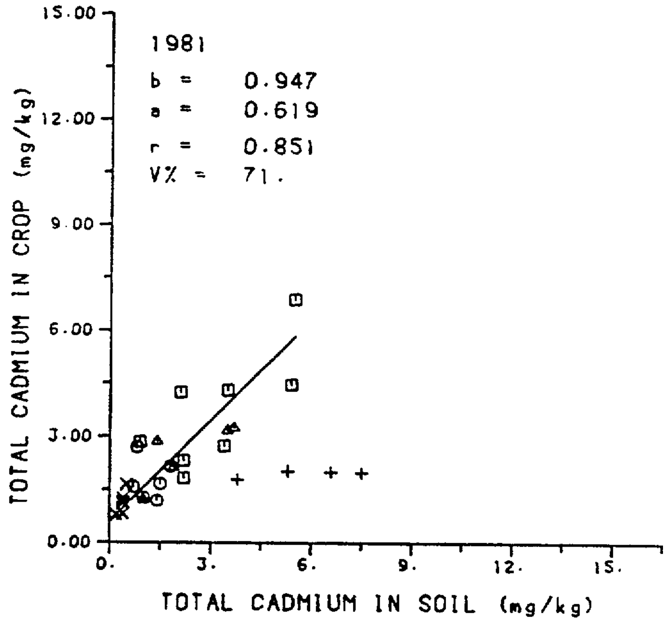
KEY

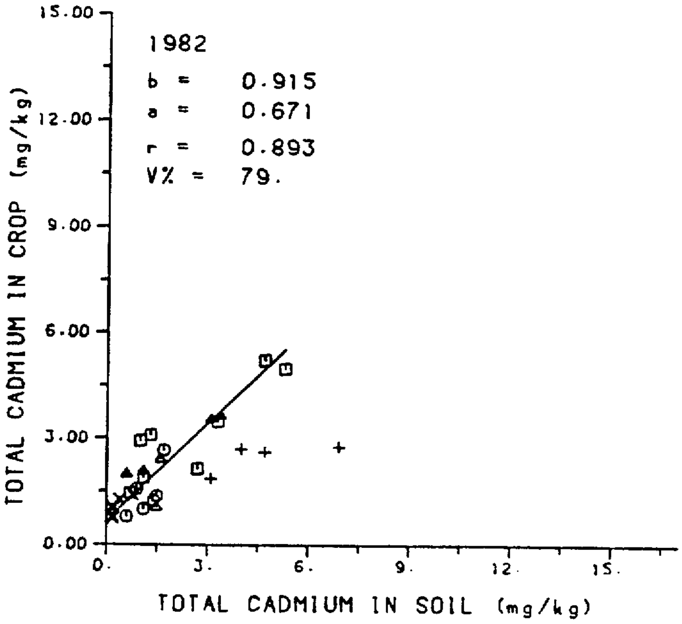
symbol	sludge type
×	background soil only
⊡	S1 Perry Oaks
⊙	S2 Hogsmill Valley
△	S3 S1/S2 mixed
+	S4 bed-dried (excl. from regression)

b slope) or crop mean
a constant) value : $\bar{y}$
r product-moment correlation coefficient
V% percent variance accounted for by the regression









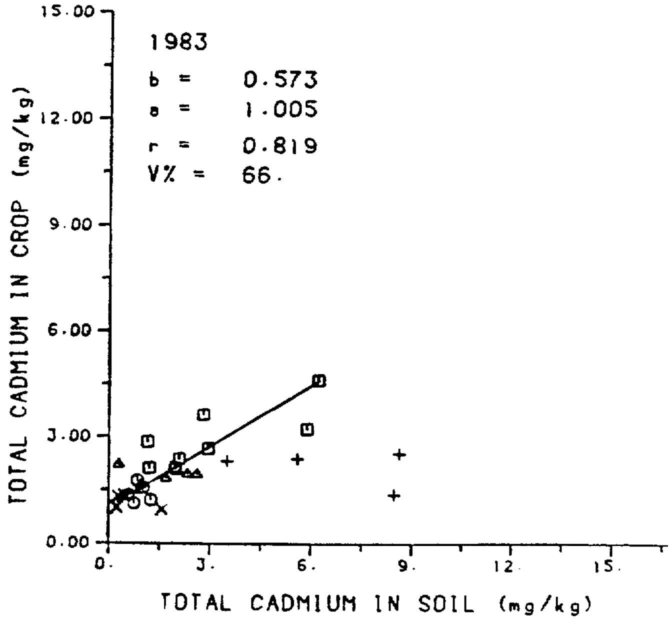


Fig K13 Cadmium in lettuce vs. total cadmium in sandy loam 1979-83

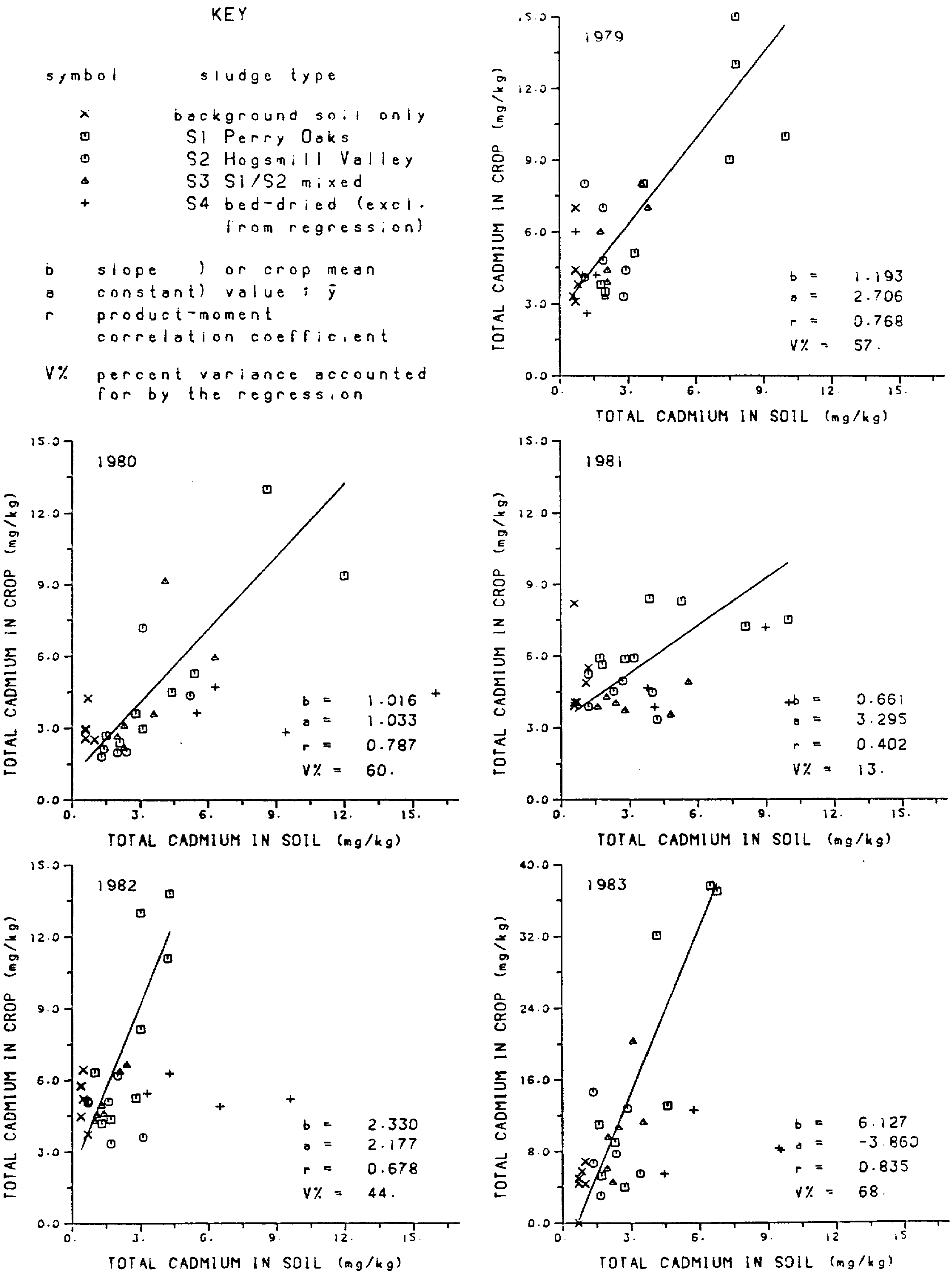


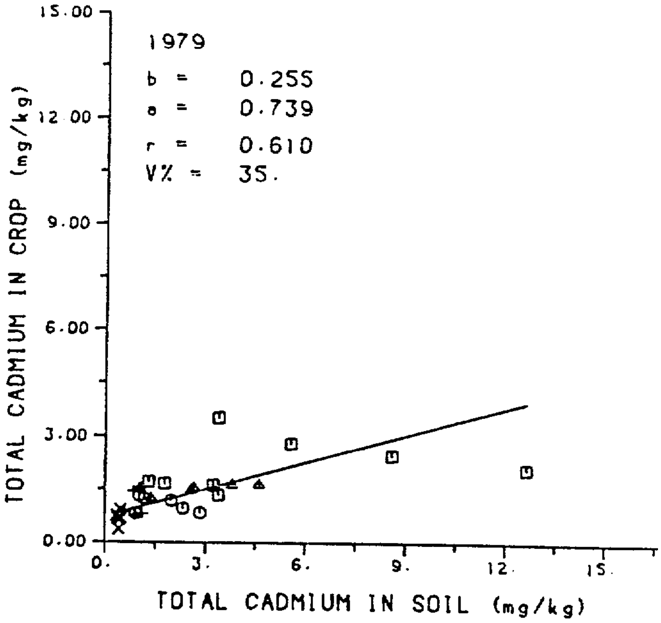
Fig K14 Cadmium in lettuce vs total cadmium in clay 1979-83

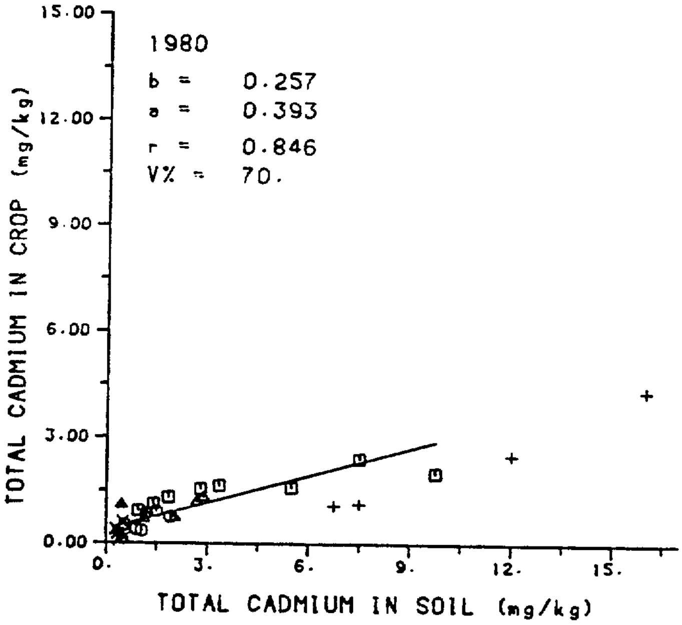
KEY

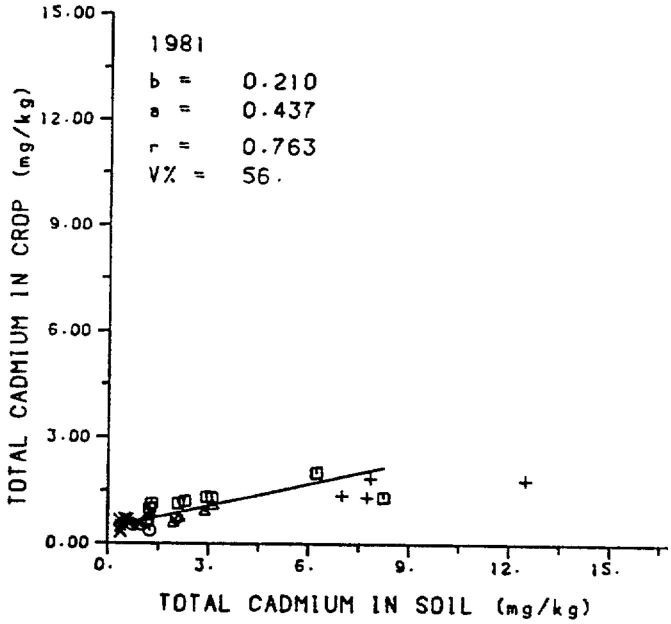
symbol	sludge type
×	background soil only
⊡	S1 Perry Oaks
ʘ	S2 Hogsmill Valley
▵	S3 S1/S2 mixed
+	S4 bed-dried (excl. from regression)

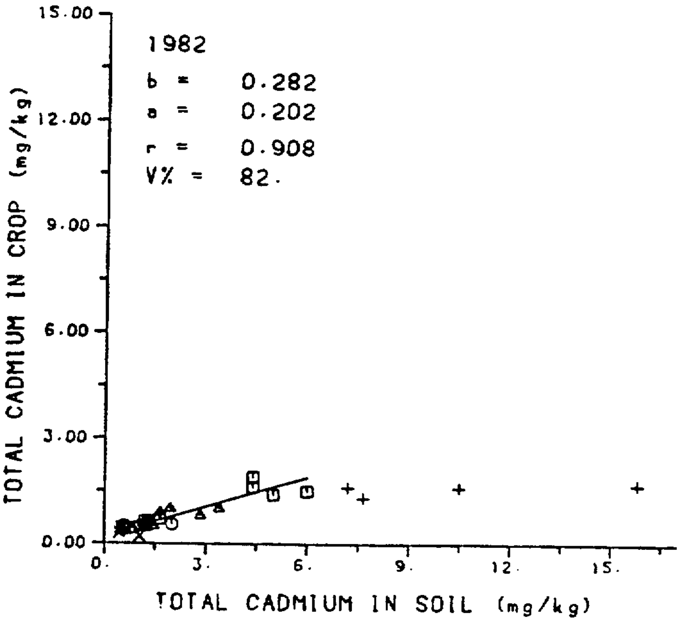
b slope) or crop mean
a constant) value : $\bar{y}$
r product-moment correlation coefficient

V% percent variance accounted for by the regression









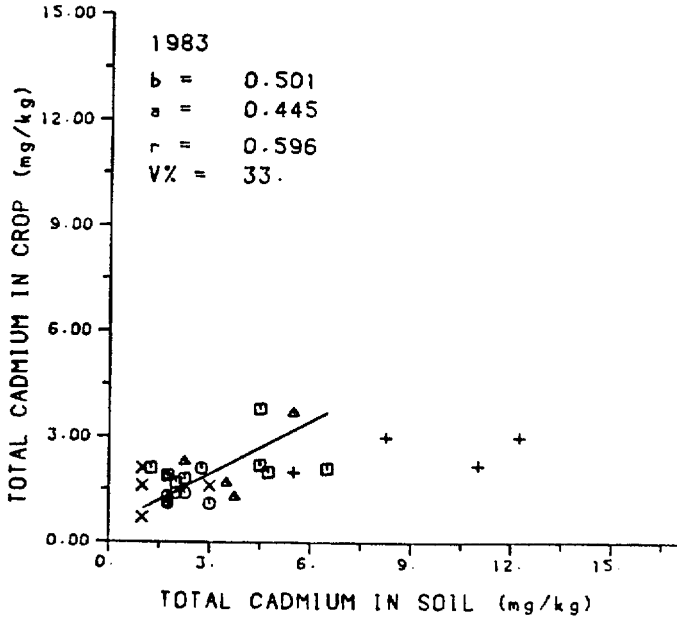


Fig K**15** Cadmium in lettuce vs. total cadmium in calc. loam 1979-83

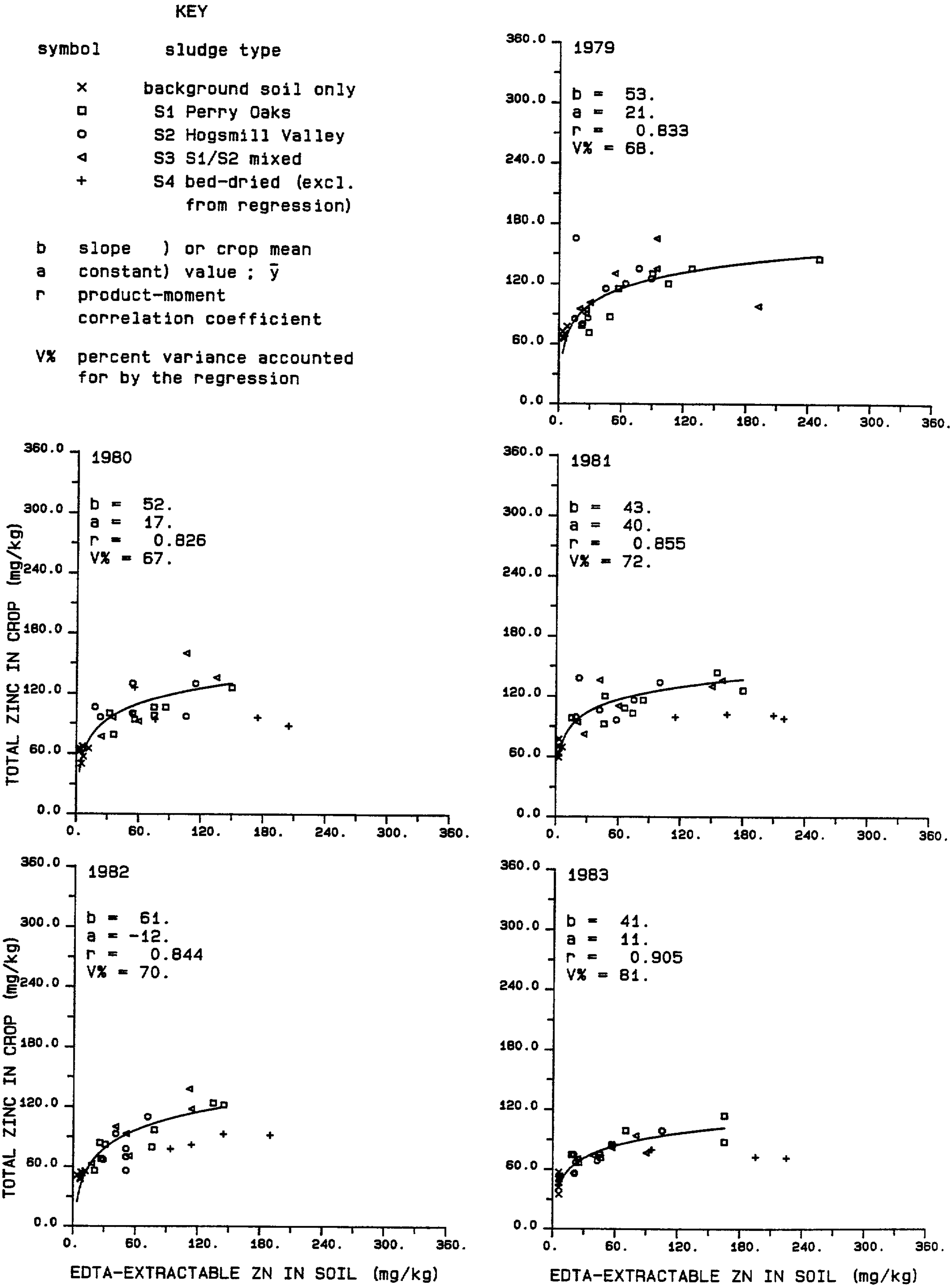


Fig K16. Zinc in lettuce vs. EDTA-Zinc in sandy loam 1979-83

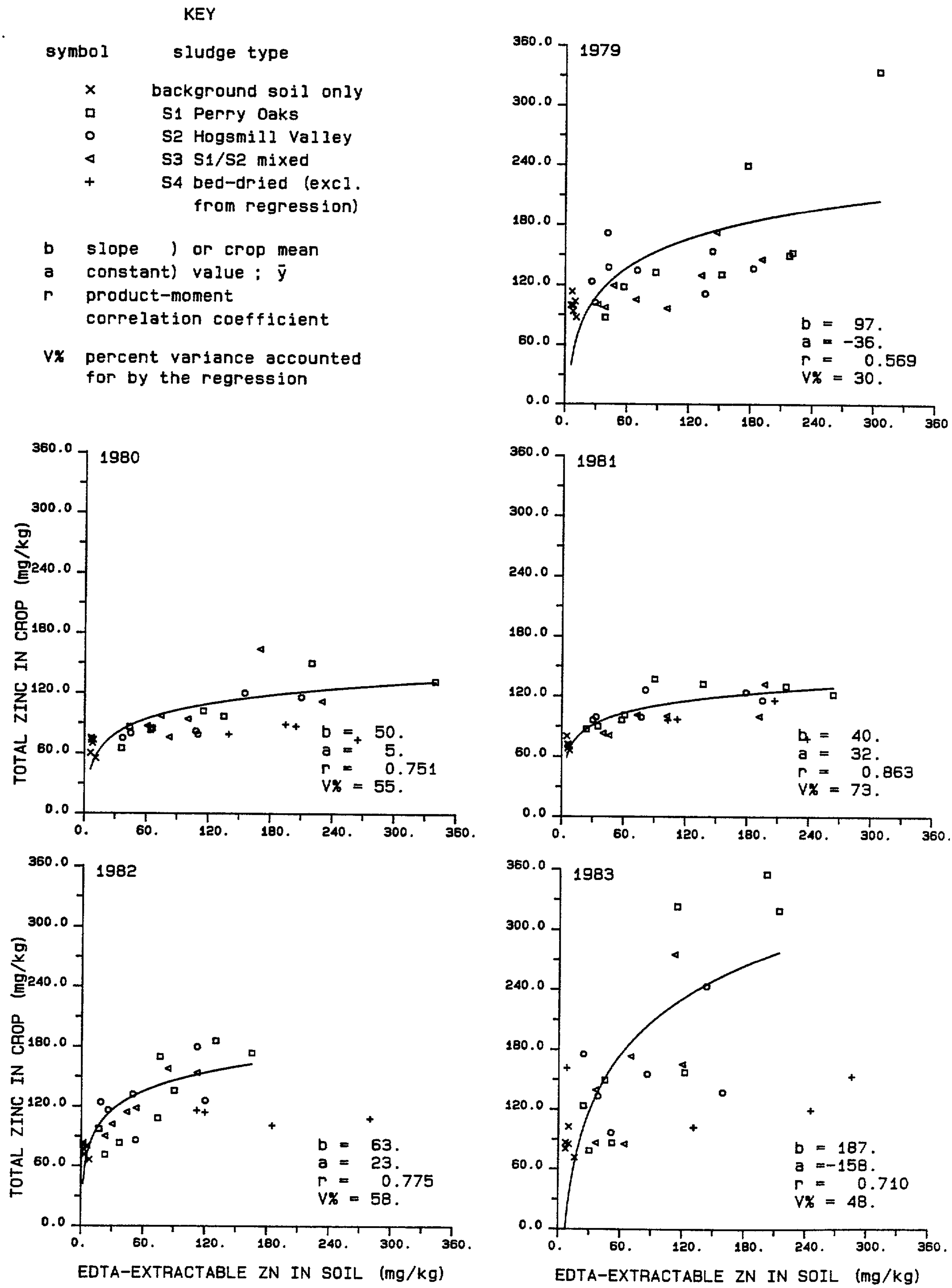


Fig K17. Zinc in lettuce vs. EDTA-Zinc in clay 1979-83

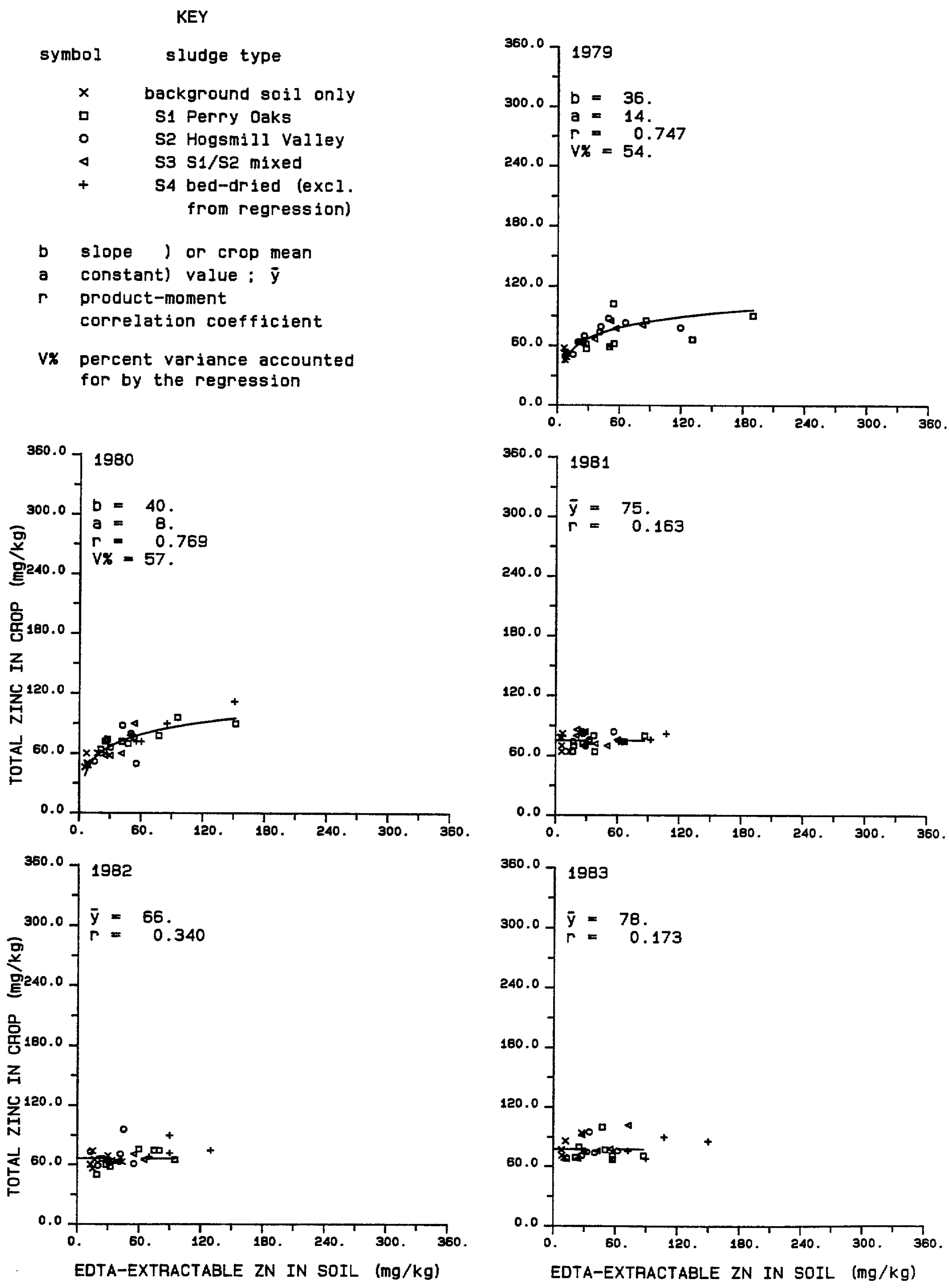


Fig K18. Zinc in lettuce vs. EDTA-Zinc in calcareous loam 1979-83

KEY

symbol	sludge type
×	background soil only
⊡	S1 Perry Oaks
⊙	S2 Hogsmill Valley
▲	S3 S1/S2 mixed
+	S4 bed-dried (excl. from regression)

b slope) or crop mean
a constant) value : ȳ
r product-moment correlation coefficient

V% percent variance accounted for by the regression

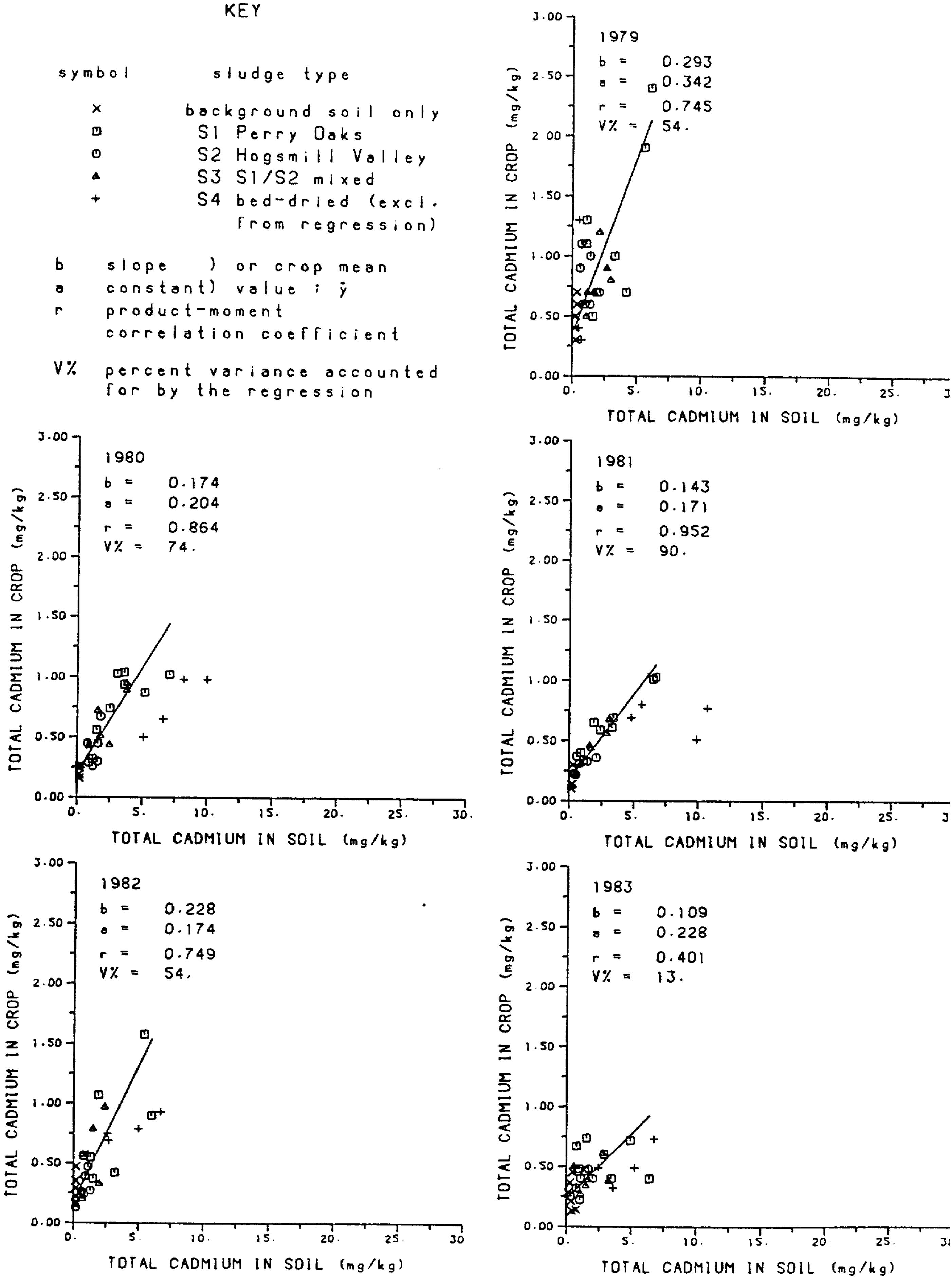


Fig K19 Cadmium in red beet vs. total cadmium in sandy loam 1979-83

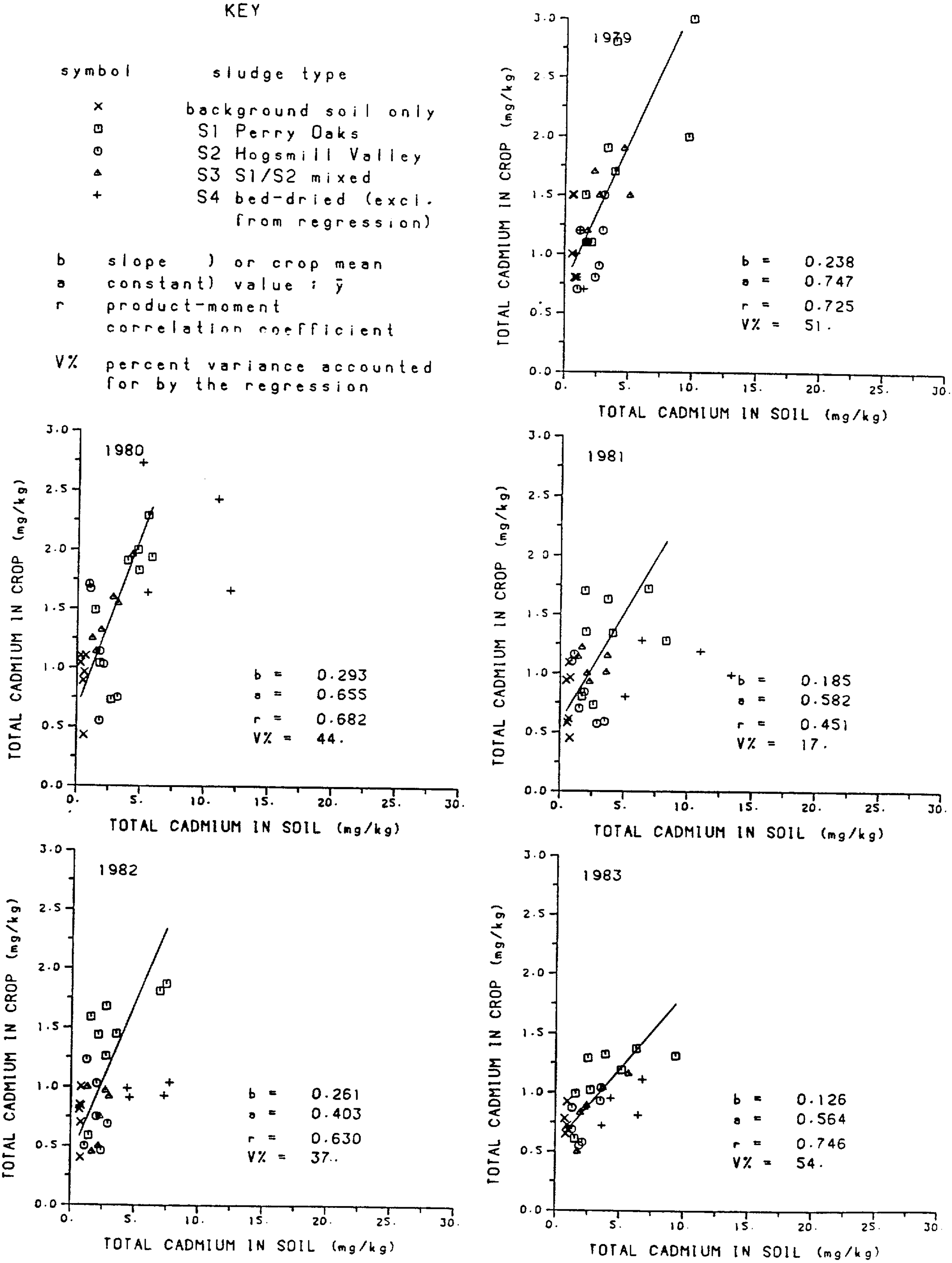


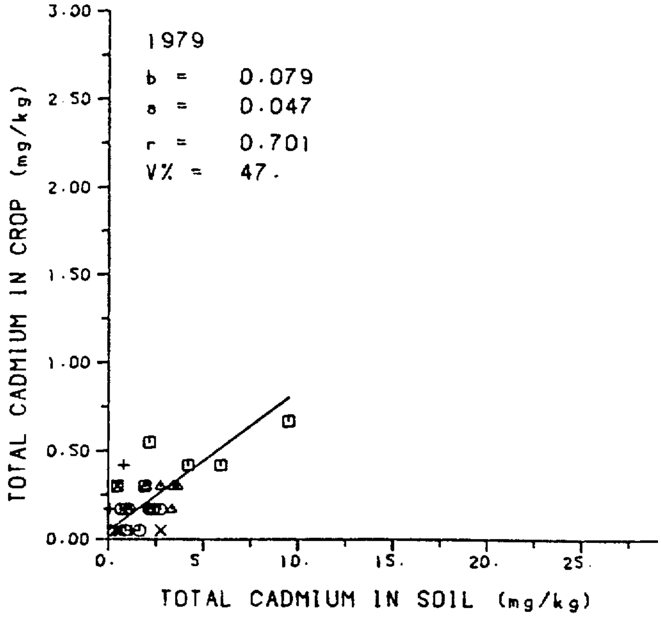
Fig K20 Cadmium in red beet vs. total cadmium in clay 1979-83

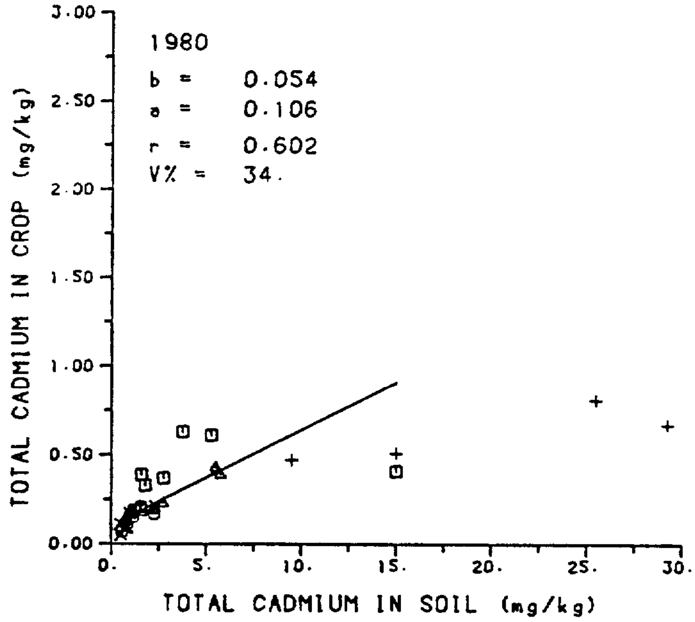
KEY

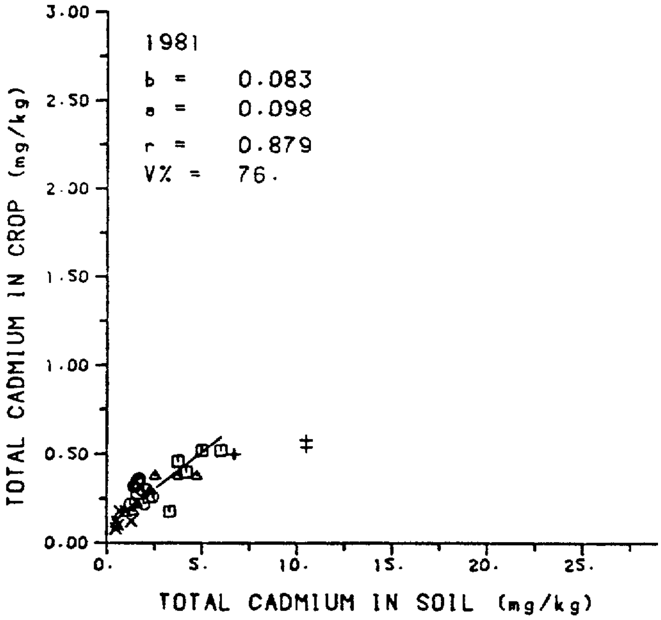
symbol	sludge type
×	background soil only
⊡	S1 Perry Oaks
⊙	S2 Hogsmill Valley
▲	S3 S1/S2 mixed
+	S4 bed-dried (excl. from regression)

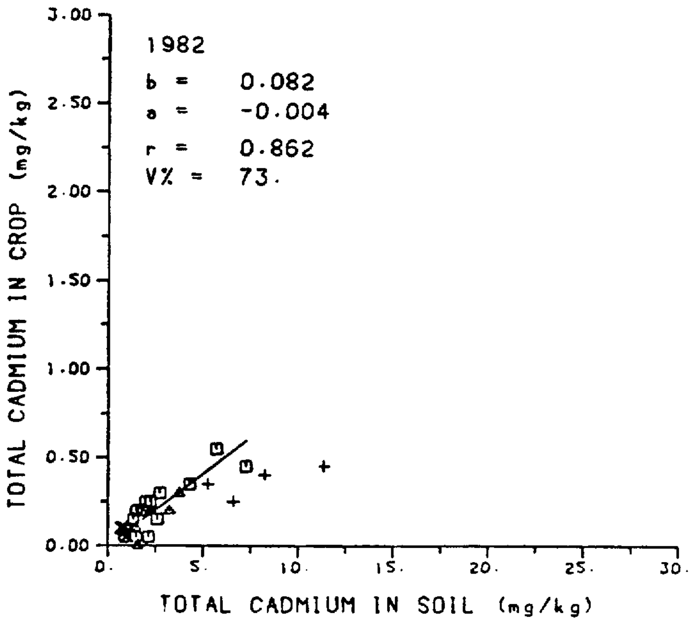
b slope) or crop mean
a constant) value ; $\bar{y}$
r product-moment correlation coefficient

V% percent variance accounted for by the regression









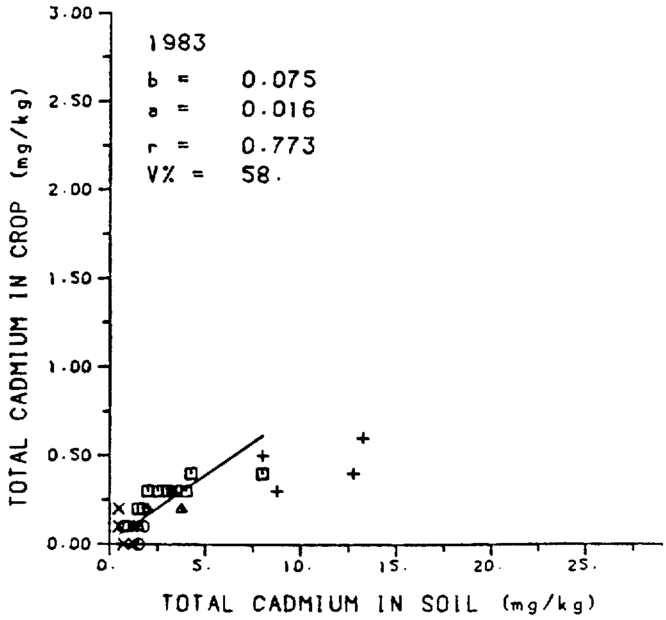


Fig K**21** Cadmium in red beet vs. total cadmium in calc. loam 1979-83

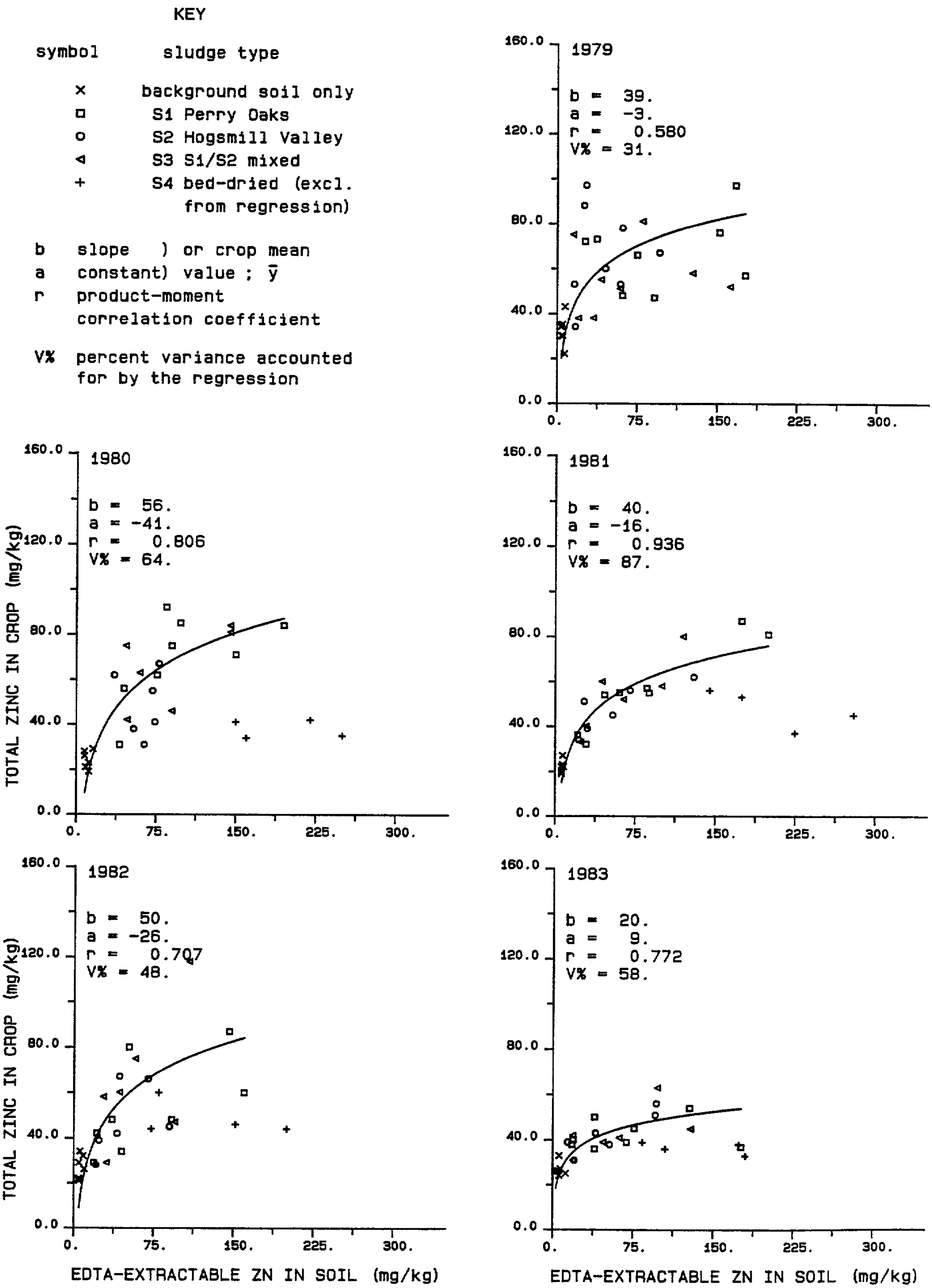


Fig K22. Zinc in red beet vs. EDTA-Zinc in sandy loam 1979-83

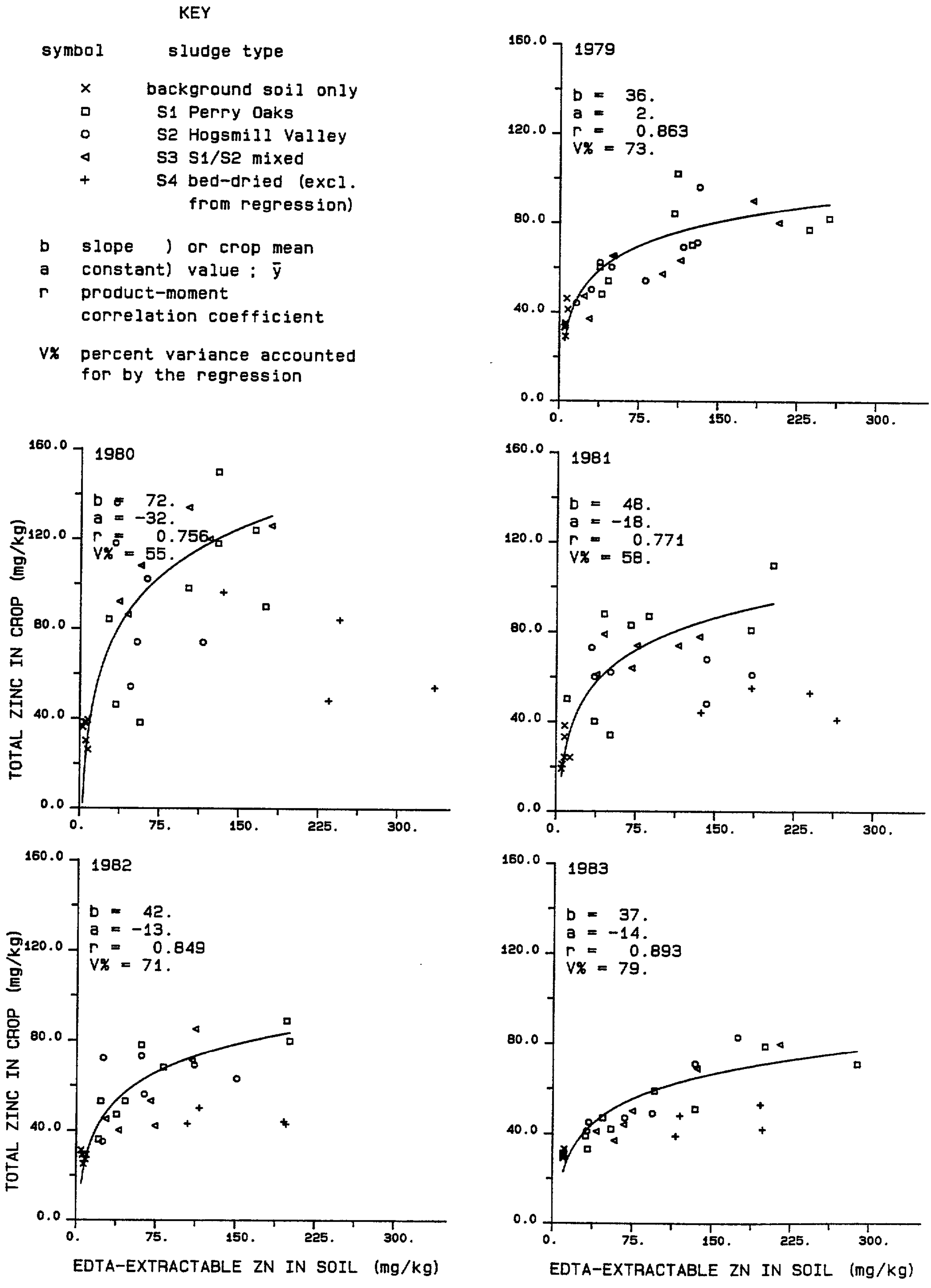


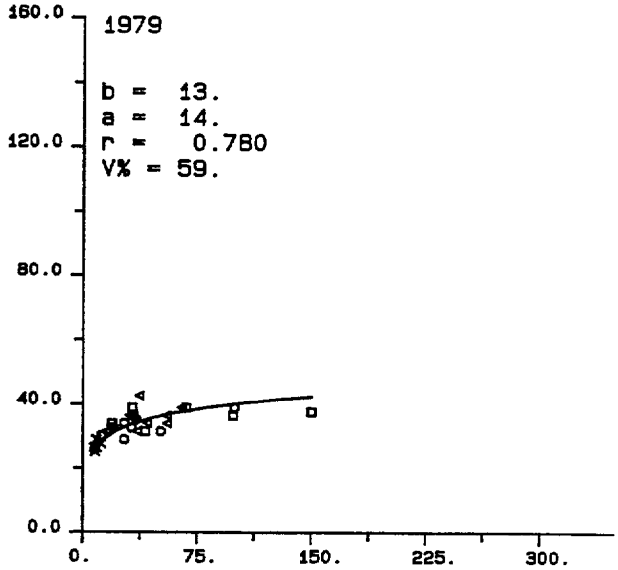
Fig K23. Zinc in red beet vs. EDTA-Zinc in clay 1979-83

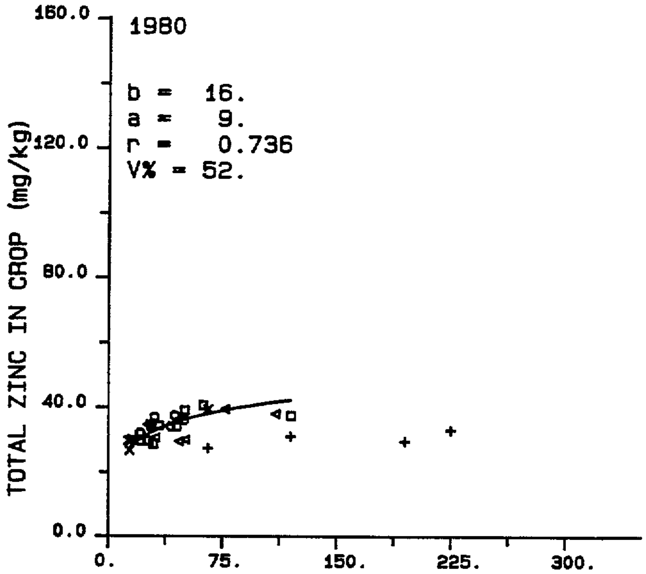
KEY

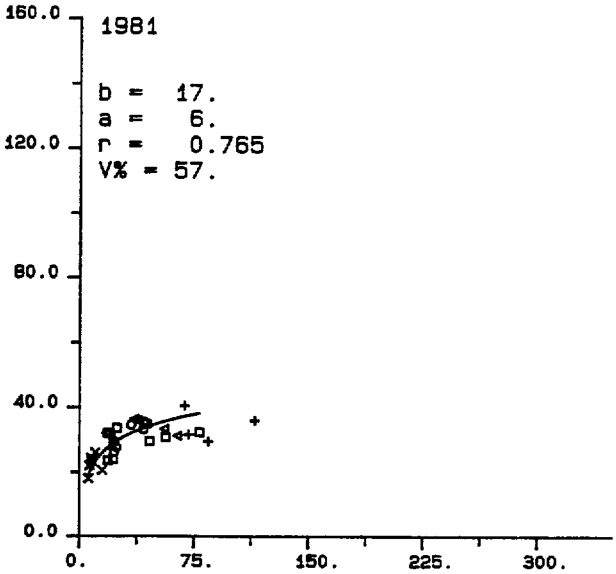
symbol	sludge type
×	background soil only
□	S1 Perry Oaks
o	S2 Hogsmill Valley
◁	S3 S1/S2 mixed
+	S4 bed-dried (excl. from regression)

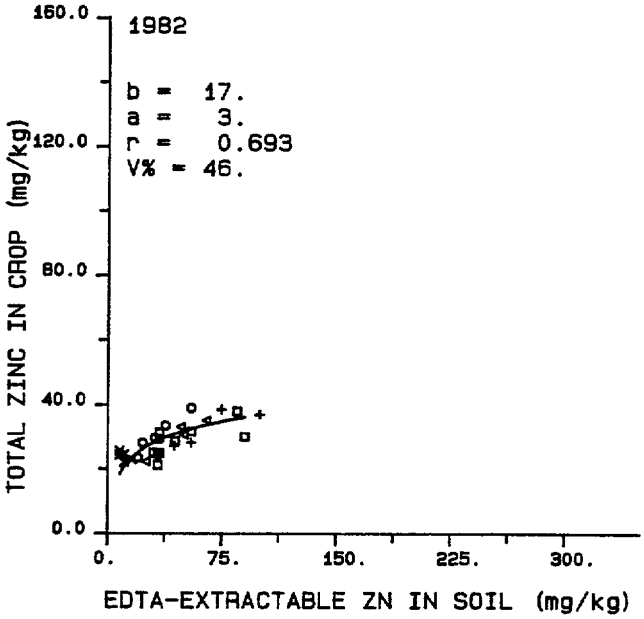
b slope) or crop mean
a constant) value ; $\bar{y}$
r product-moment correlation coefficient

V% percent variance accounted for by the regression









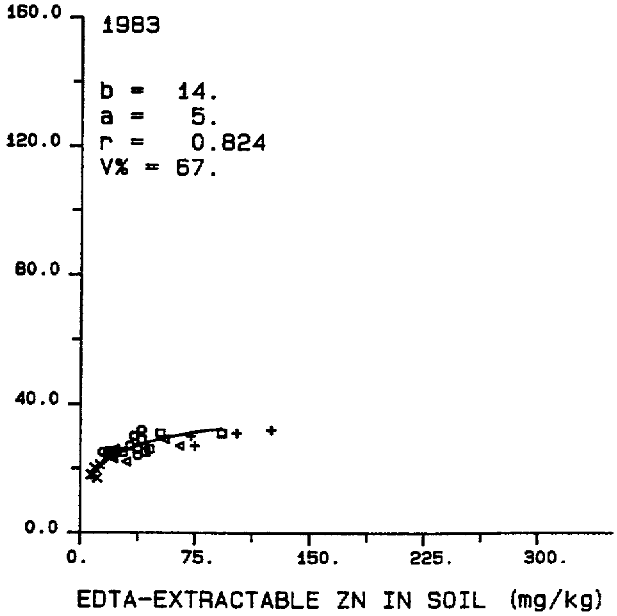


Fig K24. Zinc in red beet vs. EDTA-Zinc in calcareous loam 1979-83

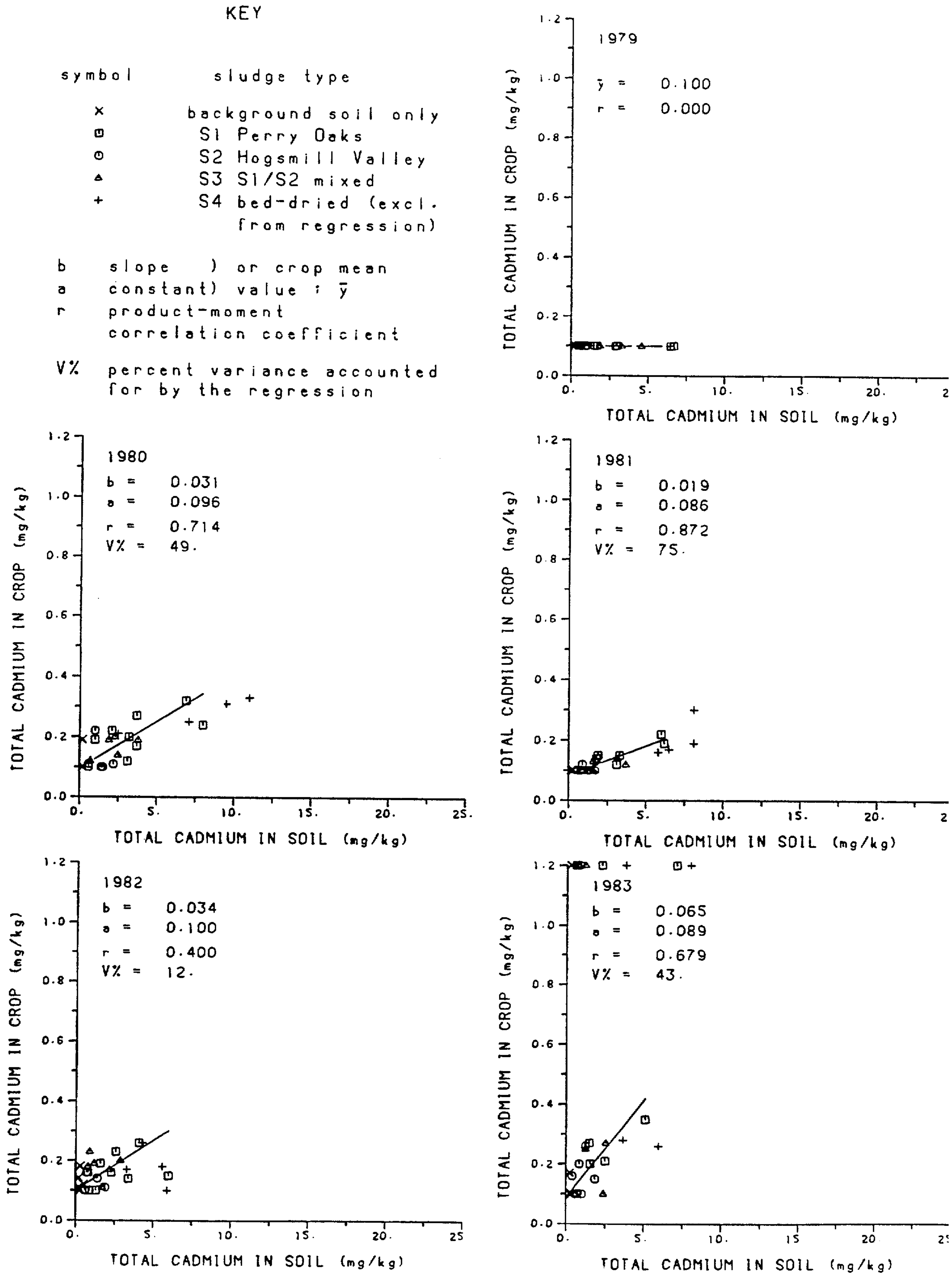


Fig K25 Cadmium in cabbage vs. total cadmium in sandy loam 1979-83

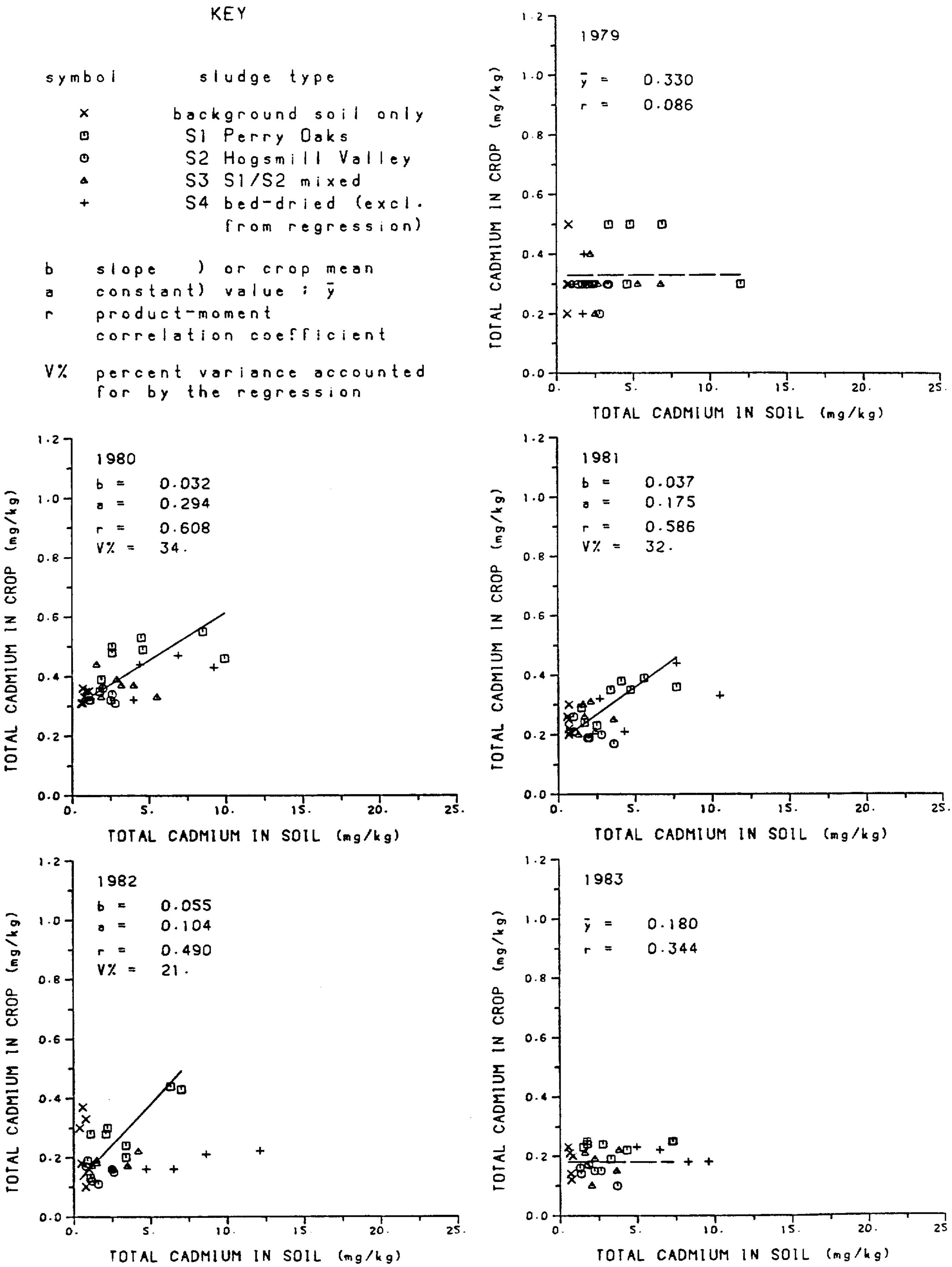


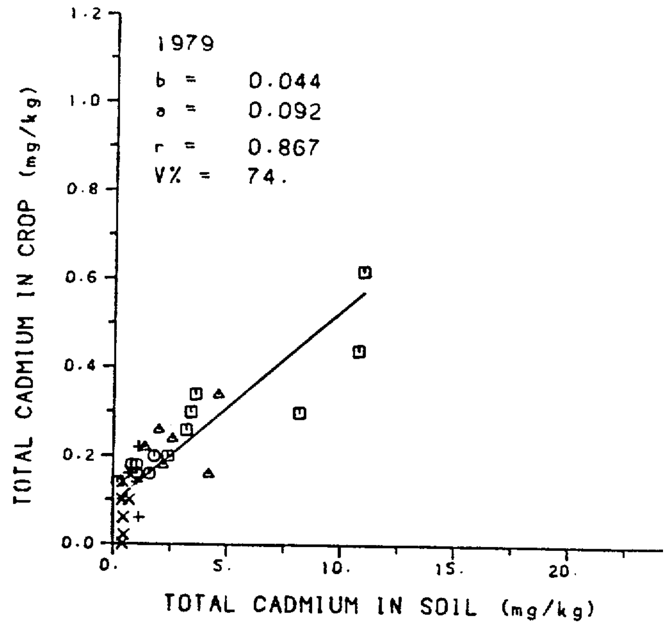
Fig K26 Cadmium in cabbage vs. total cadmium in clay 1979-83

KEY

symbol	sludge type
×	background soil only
⊡	S1 Perry Oaks
⊙	S2 Hogsmill Valley
△	S3 S1/S2 mixed
+	S4 bed-dried (excl. from regression)

b slope) or crop mean
a constant) value : $\bar{y}$
r product-moment correlation coefficient

V% percent variance accounted for by the regression



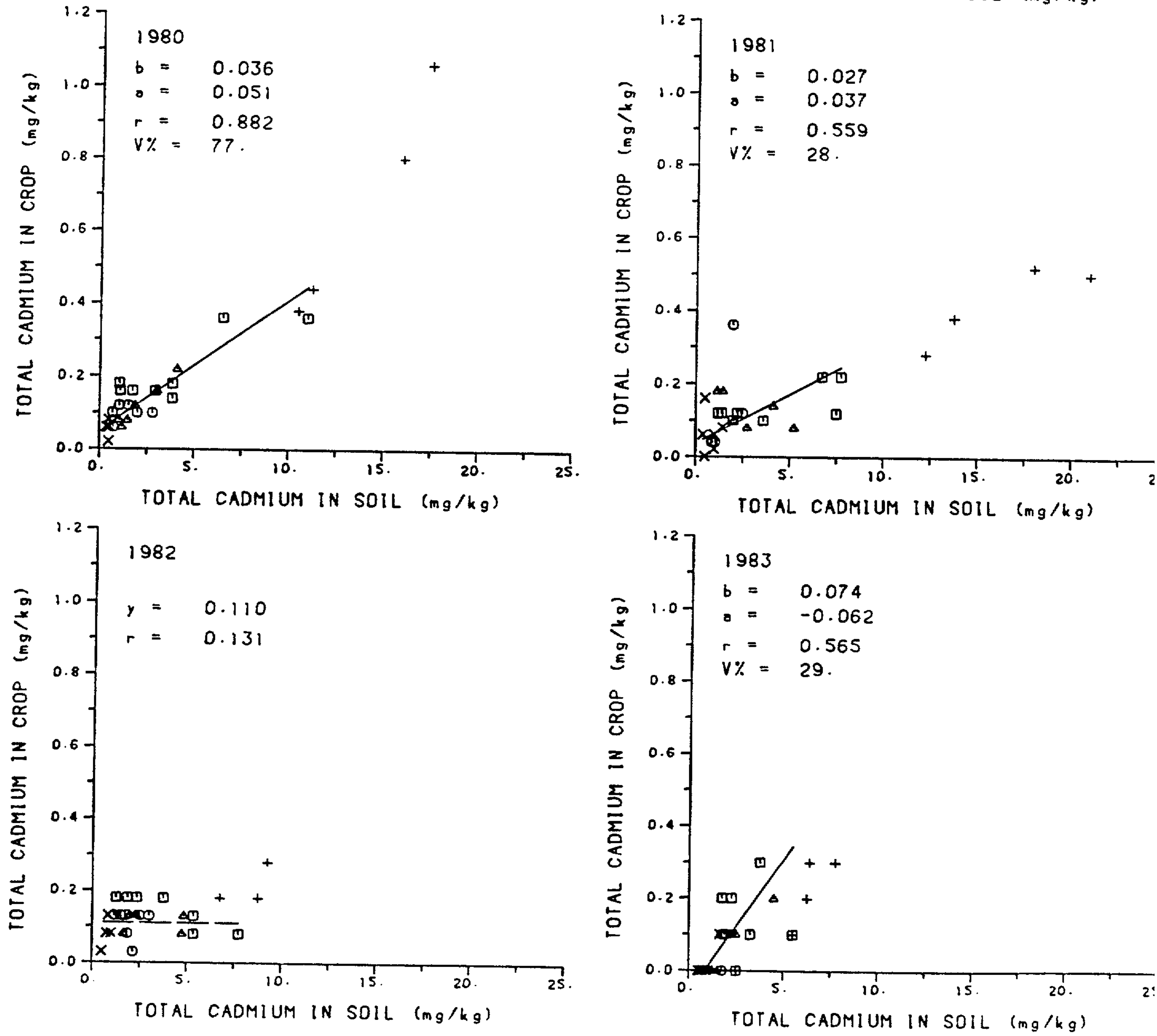


Fig K27 Cadmium in cabbage vs. total cadmium in calc. loam 1979-83

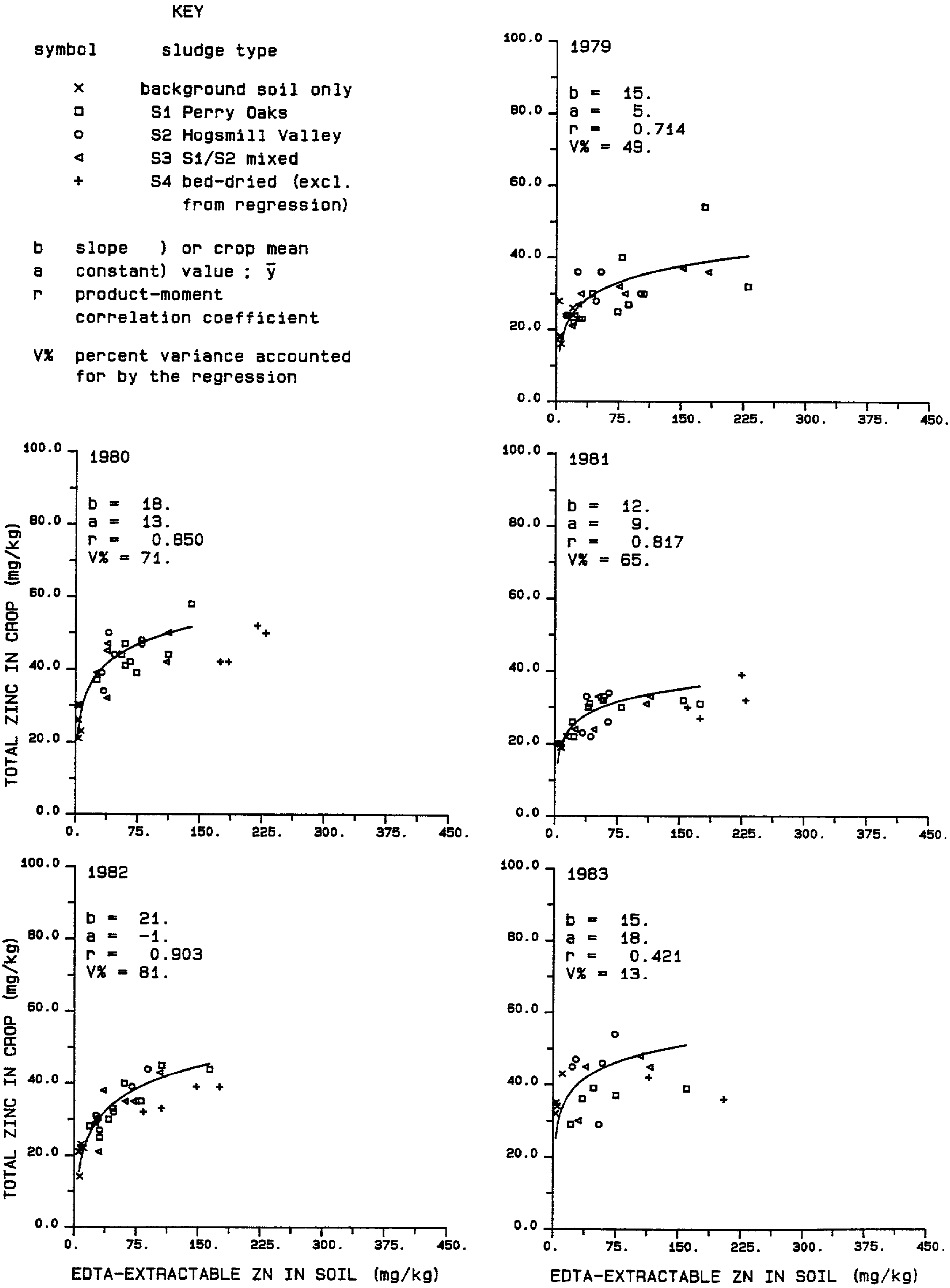


Fig K28. Zinc in cabbage vs. EDTA-Zinc in sandy loam 1979-83

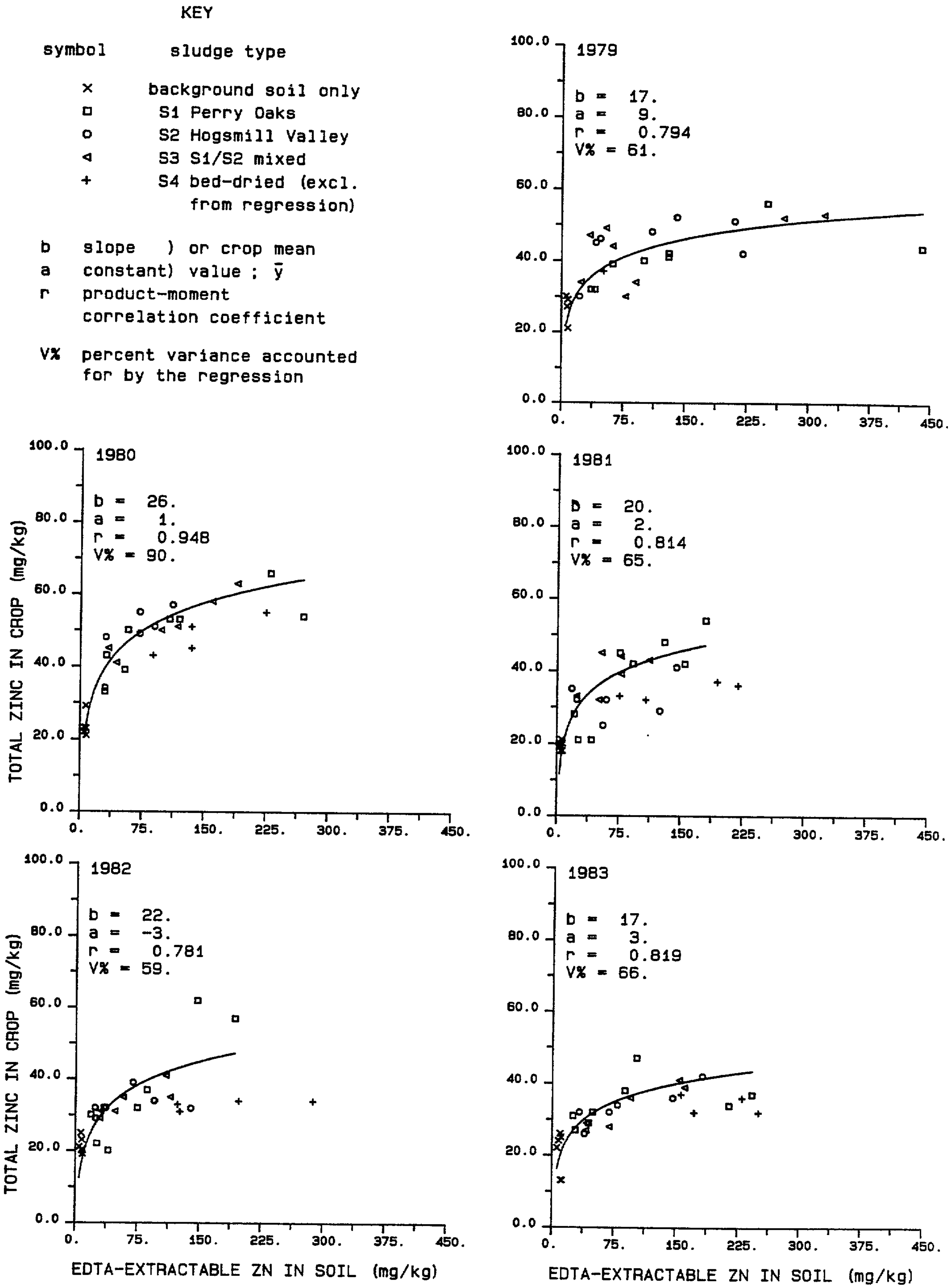


Fig K29. Zinc in cabbage vs. EDTA-Zinc in clay 1979-83

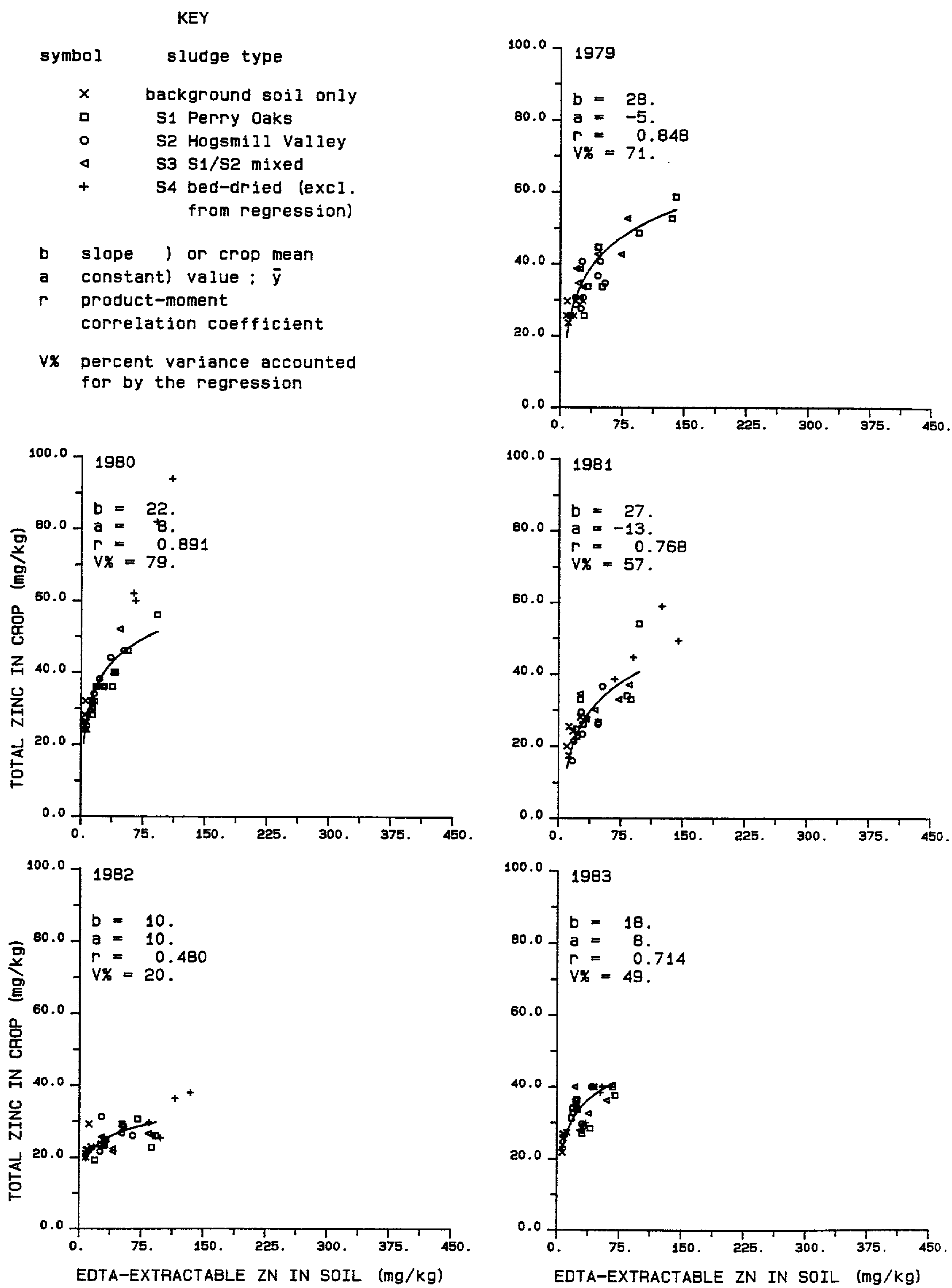


Fig K30. Zinc in cabbage vs. EDTA-Zinc in calcareous loam 1979-83

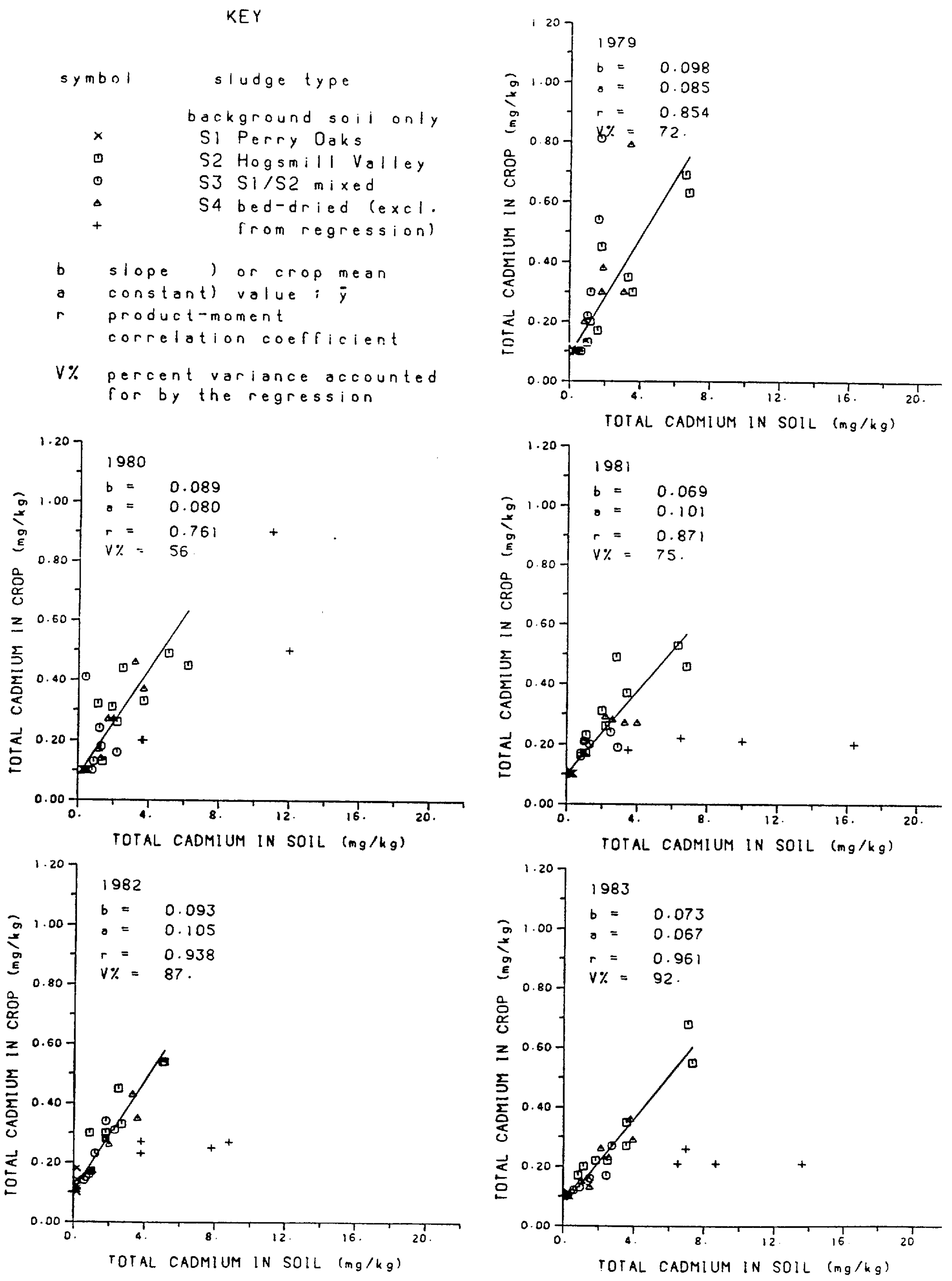


Fig K31 Cadmium in ryegrass vs. total cadmium in sandy loam 1979-83

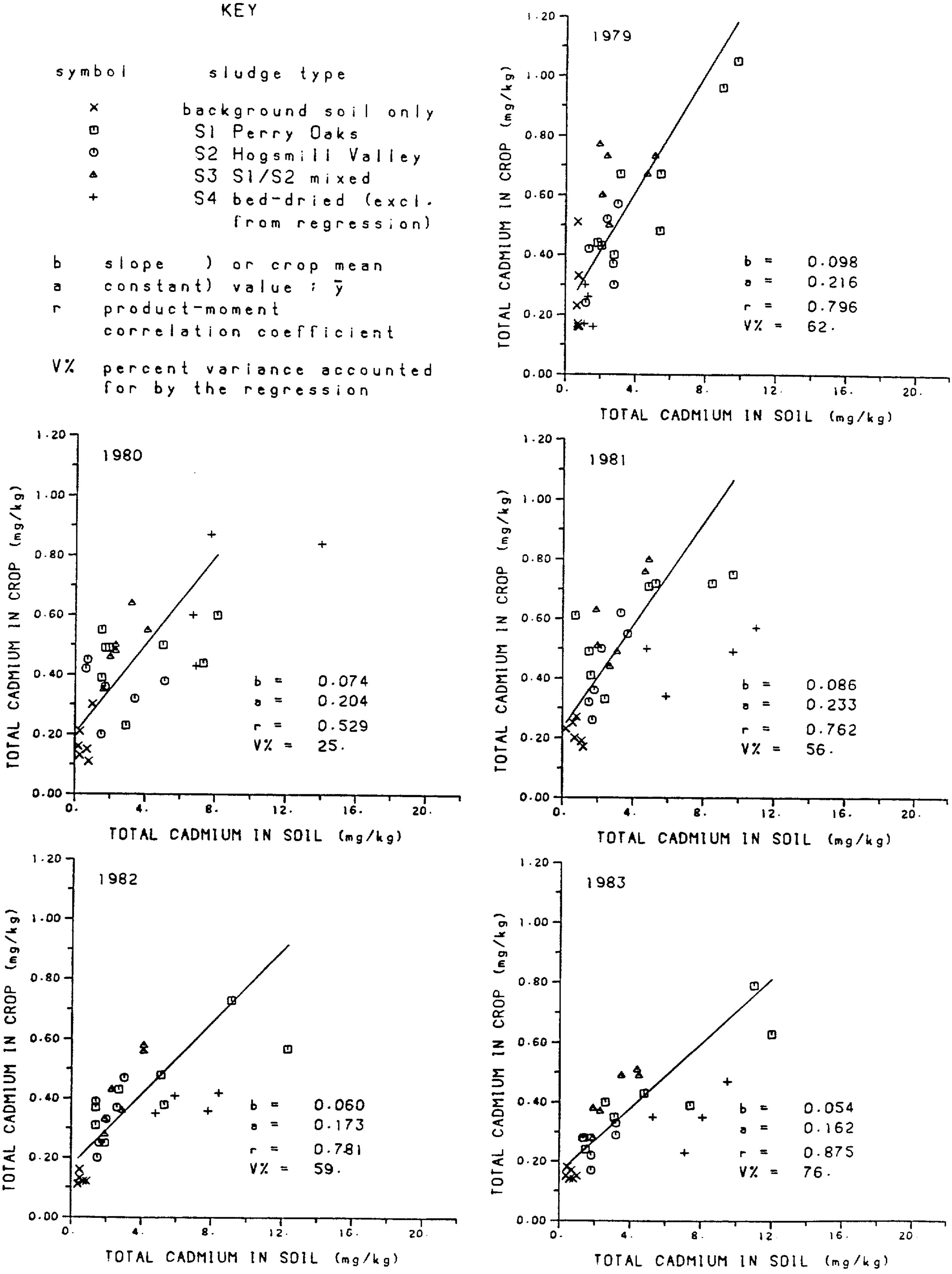


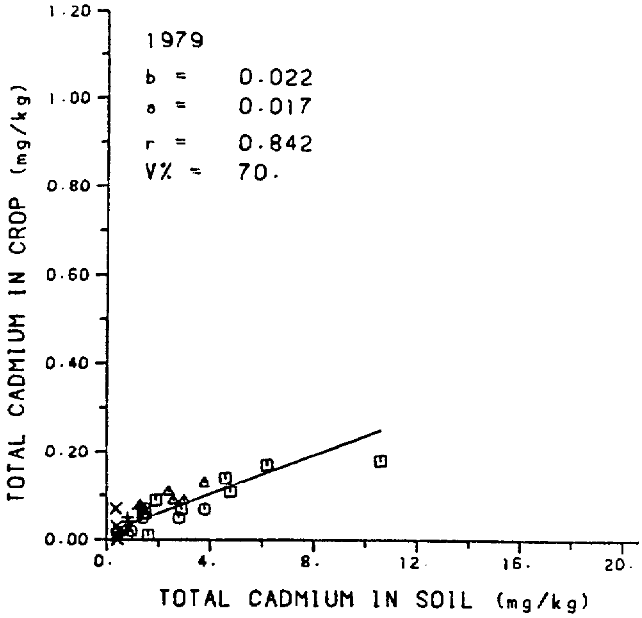
Fig K32 Cadmium in ryegrass vs. total cadmium in clay 1979-83

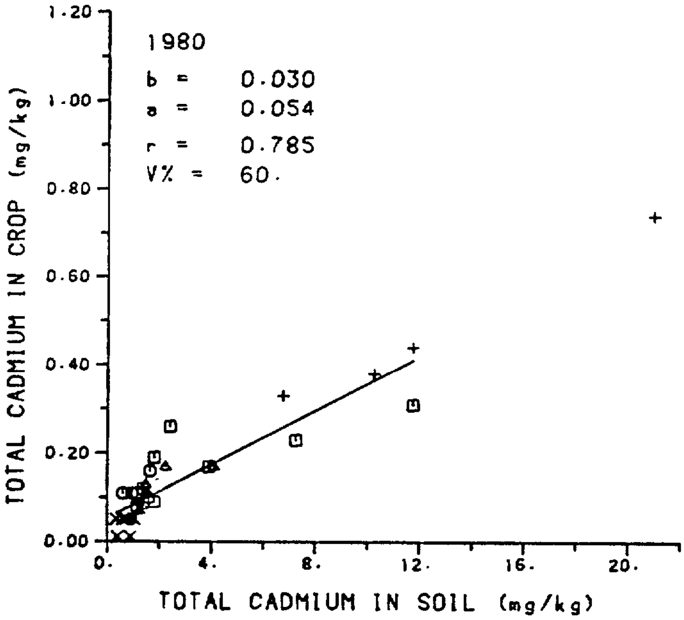
KEY

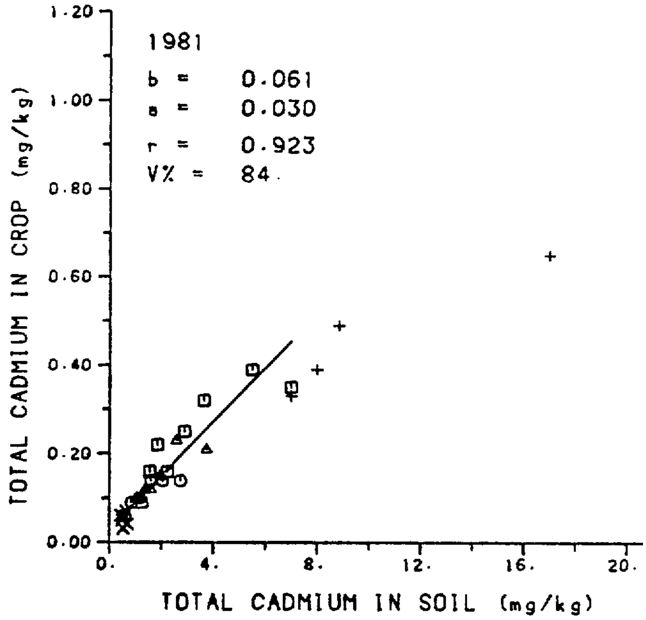
symbol	sludge type
	background soil only
×	S1 Perry Oaks
⊡	S2 Hogsmill Valley
⊙	S3 S1/S2 mixed
▲	S4 bed-dried (excl.
+	from regression)

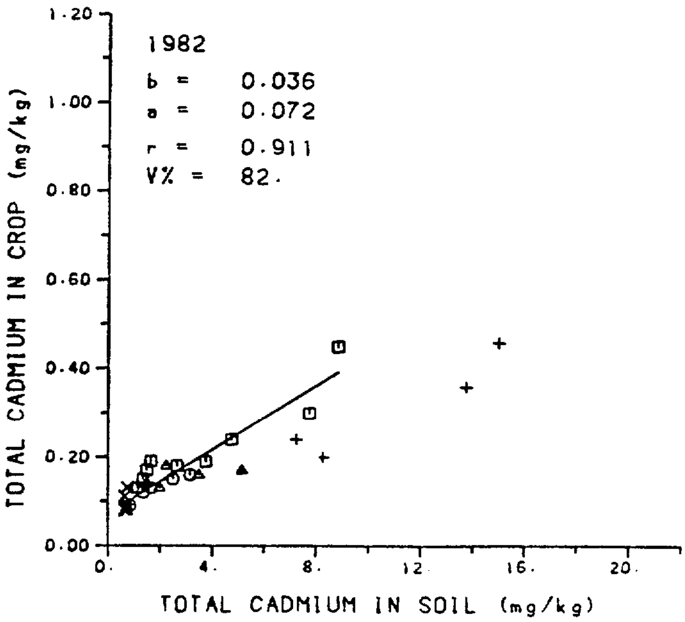
b slope) or crop mean
a constant) value : $\bar{y}$
r product-moment correlation coefficient

V% percent variance accounted for by the regression









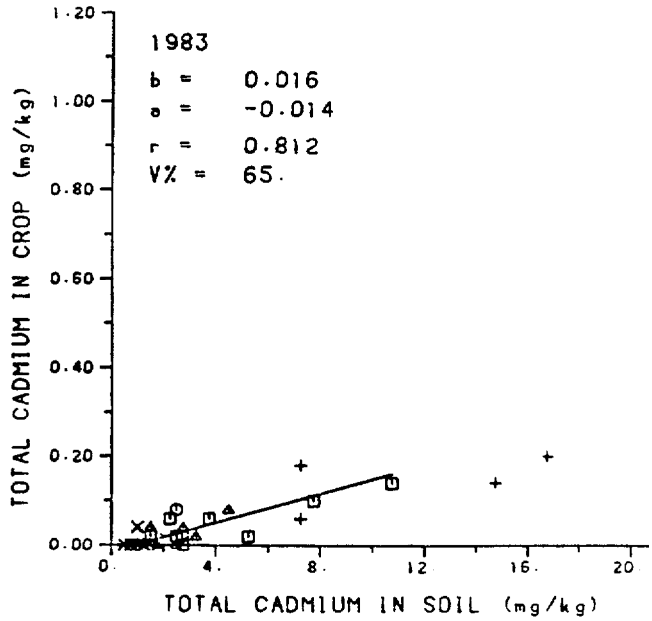


Fig K33 Cadmium in ryegrass vs. total cadmium in calc. loam 1979-83

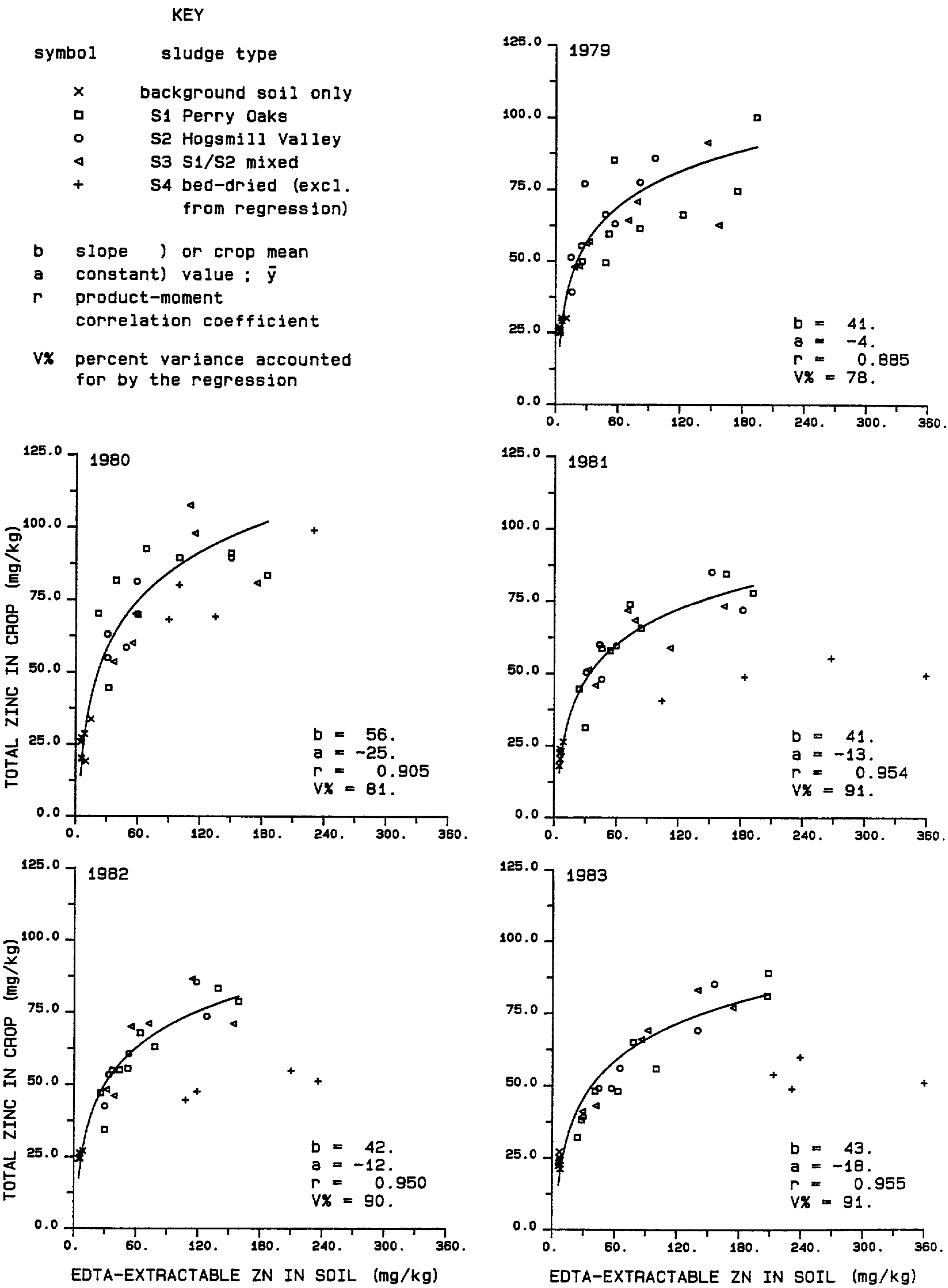


Fig K34. Zinc in ryegrass vs. EDTA-Zinc in sandy loam 1979-83

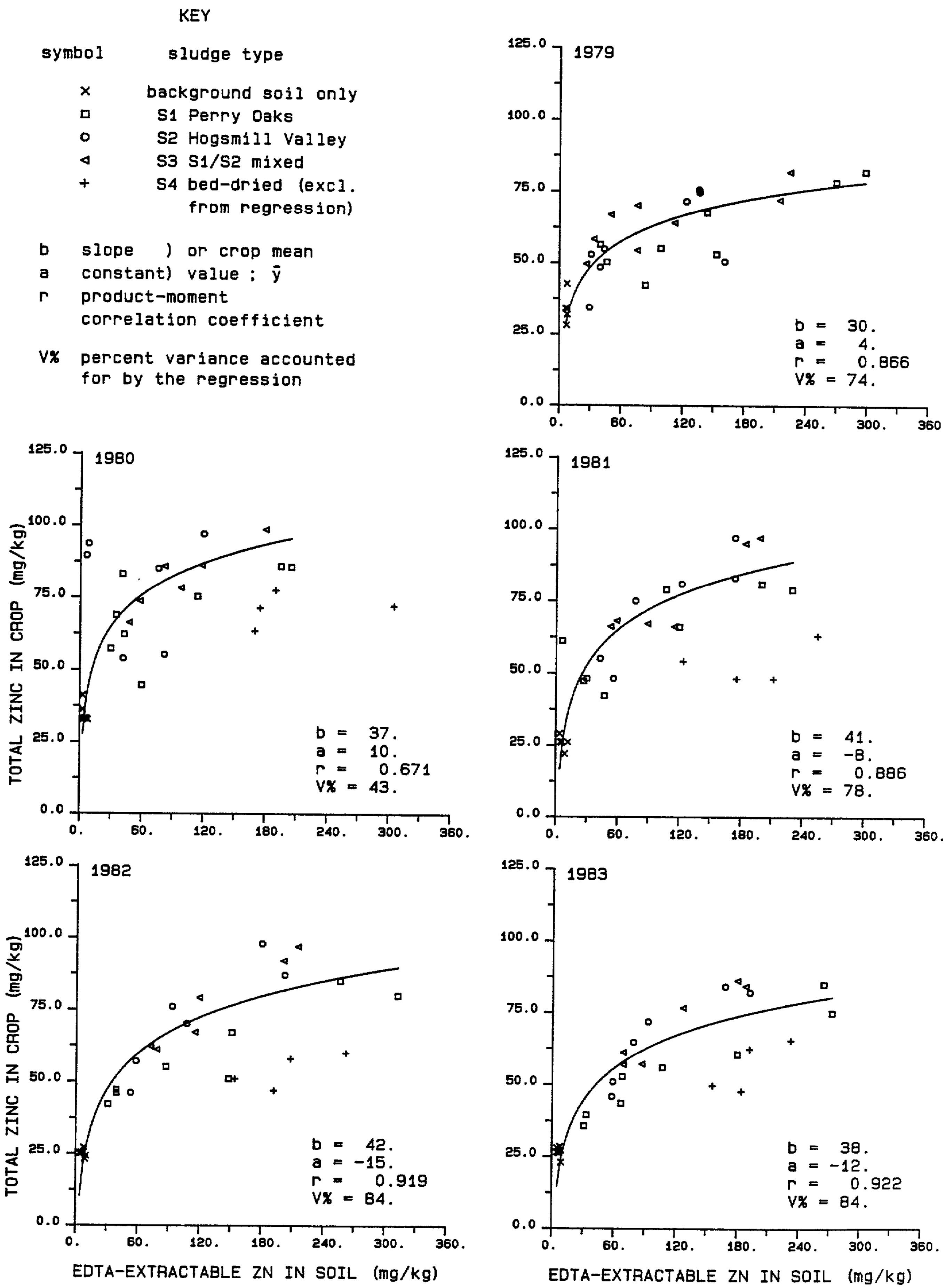


Fig K35. Zinc in ryegrass vs. EDTA-Zinc in clay 1979-83

KEY

symbol	sludge type
×	background soil only
□	S1 Perry Oaks
o	S2 Hogsmill Valley
◁	S3 S1/S2 mixed
+	S4 bed-dried (excl. from regression)

b slope) or crop mean
a constant) value ; $\bar{y}$
r product-moment correlation coefficient

V% percent variance accounted for by the regression

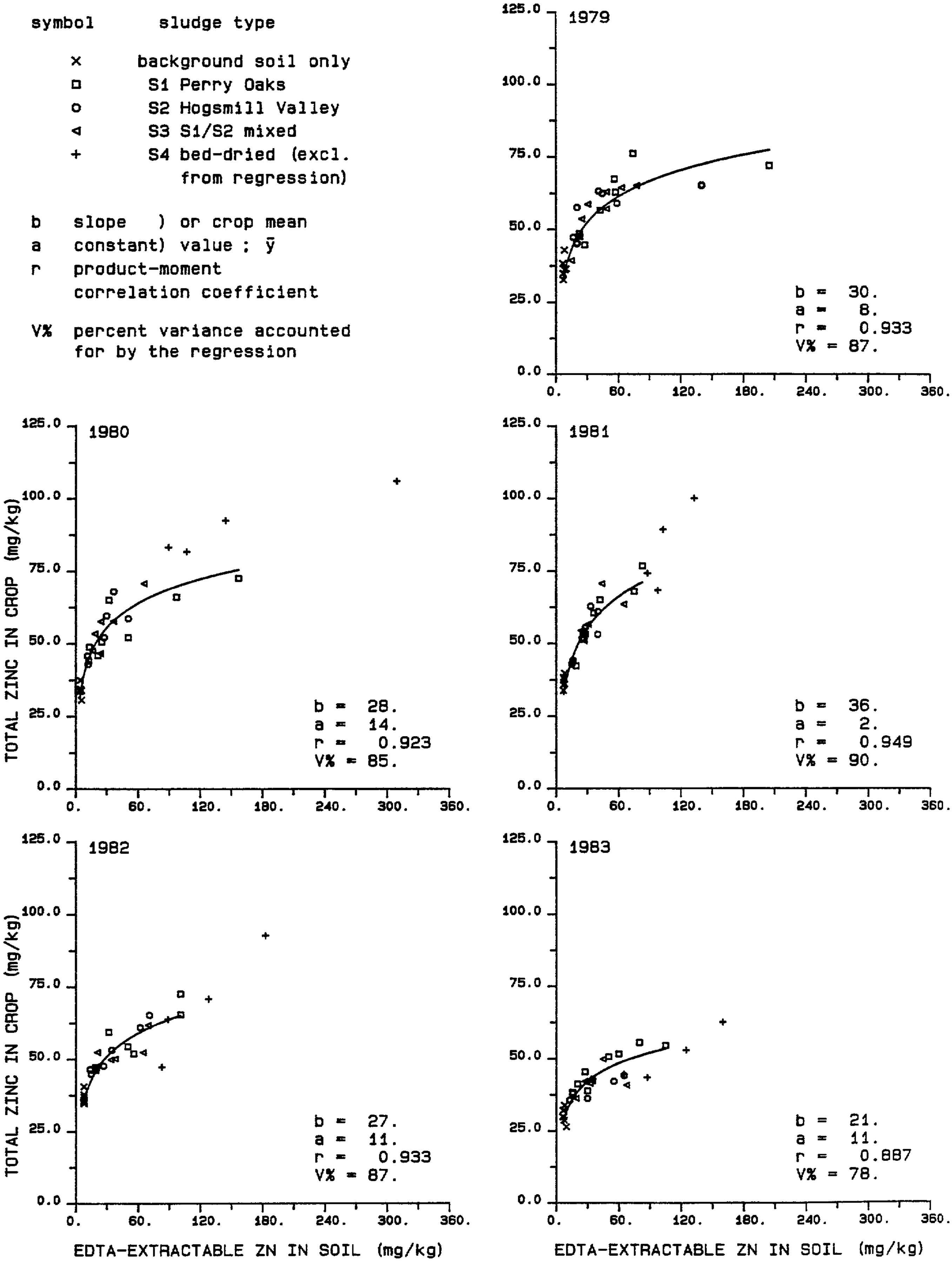


Fig K36. Zinc in ryegrass vs. EDTA-Zinc in calcareous loam 1979-83

WRc ENGINEERING
P O Box 85
Frankland Road
Blagrove, Swindon
Wilts SN5 8YR
Tel: Swindon (0793) 488301
Telex: 449541

WRc ENVIRONMENT
Medmenham Laboratory
Henley Road, Medmenham
P O Box 16 Marlow
Bucks SL7 2HD
Tel: Henley (0491) 571531
Telex: 848632

WRc (Headquarters)
John L van der Post Building
Henley Road, Medmenham
P O Box 16 Marlow
Bucks SL7 2HD
Tel: Henley (0491) 571531
Telex: 848632

WRc PROCESSES
Stevenage Laboratory
Elder Way
Stevenage, Herts
SG1 1TH
Tel: Stevenage (0438) 312444
Telex: 826168

WRc SCOTTISH OFFICE
1 Snowdon Place
Stirling FK8 2NH
Tel: Stirling (0786) 71580

WRc WATER BYELAWS ADVISORY SERVICE
660 Ajax Avenue
Slough, Bucks
SL1 4BG
Tel: Slough (0753) 37277
Telex: 449541

Registered Offices:

WRc
WRc CONTRACTS
CABLETIME INSTALLATIONS LTD
Henley Road, Medmenham
P O Box 16 Marlow
Bucks. SL7 2HD
Tel: Henley (0491) 571531
Telex: 848632