Studies on the Uptake of Soil-Applied Mercury

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Public attention in Canada was focussed in the fall of 1969 on the possible hazardous effects of mercury entering the food chain, when it was reported that fish taken from the South Saskatchewan River contained up to 10 ppm mercury. Around the same time the hunting of pheasants and Hungarian partridges were temporarily banned in Alberta, after unusually high levels of mercury were reported in the muscle tissue of these birds. The setting up of a national survey to determine the extent and degree of mercury contamination of Canadian fish resulted in a banning in March 1970, of the sale and export of fish taken from the Canadian waters of Lake St. Clair.

Although mercury has been used in Canada for over 40 years in the treatment of seed against important fungal diseases, there is no evidence to suggest that this necessary and effective practice has directly contributed to the mercury pollution problem in Canada. Concern regarding the use of mercury as a seed treatment is not from the addition of small amounts of mercury to the soil (Table 1), or possible uptake and translocation of mercury in the plant, but rather problems related to the use of mercury fungicides including:

1. direct uptake of mercury treated grain by seed eating birds and rodents;
2. human consumption of mercury treated grain;
3. feeding (accidental or otherwise) of mercury treated grain to livestock; and
4. contamination of animal feeds from seed bins, truck boxes and grain augers that were used to handle mercury dressed grain.

Because of the importance of relatively low levels of mercury in the aquatic environment, this subject has received much attention in recent years. Little information is available on soil-mercury-plant relationships. This present study was initiated to determine mercury levels in agriculturally important soils and crops in Saskatchewan, and provide data to help clarify the following relationship:

Hg (parent material) $\rightleftharpoons$ Hg (soil) $\rightleftharpoons$ Hg (roots) $\rightleftharpoons$ Hg (plant stems and leaves, seeds and roots)

Table 1

Addition of mercury to the soil as mercury seed dressings

e.g. Wheat var. Manitou dressed with 5% Hg fungicide

Seed rate - 1 bushel/acre

Hg application - $\frac{1}{2}$ oz. of 5% Hg fungicide/bushel

i.e. $\frac{1}{2}$ oz. of 5% Hg/acre

or 1/40 oz. of Hg/acre

1/40 oz. Hg in 2 million pounds

1/(40 x 2 x 16) pounds in 1 million pounds

i.e. < .001 p.p.m.

Mercury in Soils

The abundance of mercury in topsoil has been measured in various regions of the world. As early as 1934, Stock and Cucuel proposed a mean value of 70 ng Hg/g for all soils. Andersson (1), analyzed both Swedish and African topsoils and found the mean mercury contents to be 60.1 and 23 ng Hg/g of soil respectively. Levels in soils gathered throughout the states of Texas and Michigan were reported by Melton, Hoover and Howard (11) to range from 16 to 160 ng Hg/g except for a black clay which contained 1680 ng Hg/g. Martin (11) gives the natural mercury content of some English soils as between 12 and 50 ng Hg/g.

Warren and Delavault (17) measured the mercury levels in selected areas of Great Britain, and the soils were found to contain highly anomalous amounts of mercury - a few at least containing not less than between 5 and 15 μg Hg/g. The authors believe that most of the mercury in soils is of geological origin. Mercury levels in soils from British Columbia were reported by Warren, Delavault and Barakso (18). In areas referred to as unmineralized areas they found levels of 10 to 50 ng Hg/g. In soils near mineralization, (i.e. near mined or known ore bodies) levels of up to 2.5 μg Hg/g were recorded.

It appears from the literature that mercury levels in topsoil fall into two distinct categories; high levels $>$ 200 ng/g associated with areas of mercury containing parent materials such as some Precambian shales or ore bodies and much lower levels $<$ 100 ng/g. Levels in the agriculturally important soils of Saskatchewan fall into the latter category (Tables 2 and 3). Most of the soils analyzed have a mercury content of 10 to 50 ng/g, and none of the soils examined had a mercury content over 60 ng/g. As expected, there is little difference in the Hg levels of virgin and cultivated soils - as (Table 1) the use of mercury seed dressings adds little mercury to the soil; also treatment of cereal and flax seed across Canada in 1967-1968 was estimated as 40% in Saskatchewan compared to 75% in Alberta and almost 100% in Ontario (6). Wet or humid conditions favour the growth of seed disease organisms, and because of this seed treatment appears to be more common in areas where such conditions exist (2).

From the results in Tables 2 and 3, there are no distinct trends in the mercury content of the various horizons within the soil profile, except that the lowest levels were recorded in Ae horizons. This is in direct contrast to Andersson (1) who suggested that "mercury is dispersed within the soil profile so that the contents of the upper subsoil of both cultivated and untilled soils are very low, about 2-10 ng Hg/g, whereas in the topsoil these are 5 to 10 times higher". Goldschmidt (5) also considers that mercury accumulates in the upper soil horizons (Ah) which are derived from forest litter.

Table 2

Mercury content in ng/g air-dry soil of
some cultivated soils in Saskatchewan

Association	Horizon	Hg content (ng/g)
Woodmount	Ap	25
	B_1	20
	B_2	29
	Ck_1	13
Bradwell	Ap	16
	B_1	16
	Ck_1	26
Oxbow	Ap	12
	B	17
	Ck	19
Melfort	Ap	34
	B	39
	Ck	60
Waitville	Ap	28
	Bt	34
	C	23
Weyburn	Ap	36
	B_1	11
	B_2	18
	Ck_1	18

Table 3

Mercury content in ng/g air-dry soil of some virgin soils in Saskatchewan.

Association	Horizon	Hg content (ng/g)
Pelly	Ahe	23
	Bt	21
	Ck	23
Waitville	Ae	9
	Bt	30
	Ck	23
Waitville	Ae	5
	Bt	23
	Ck	19
Cypress	Ah	27
	B_1	33
	Ck_1	27
Oxbow	Ah	19
	AB	21
	B_1	17
	B_2	13
	Ck	20
Melfort	Ah	19
	B	57
	Ck	26

Mercury in Plants

Background Levels

From the literature it would appear that background levels of mercury in plant material are very small. Smart (14) states levels are often of the order of nanograms or tens of nanograms, and quotes two private communications stating that the levels in wheat and barley grain are 8 to 12 ng Hg/g whether or not they have been grown from Hg treated grain. Furutane and Osajima (1965) reported background levels in wheat, grown from undressed seed of 10 to 15 ng Hg/g. Saha et al. (1971) found no significant difference in the mercury content of wheat and barley grains whether or not they were grown from Hg treated seed. Levels ranged from 8 to 16 ng Hg/g. Also there was no significant difference at the 5% level in the results presented by James, Lagerwerff and Duffy (7), where wheat was grown from Hg treated and untreated seeds at the pH's 5.6 and 7.1. Finally John (1972) gives the background mercury level of oat grain as 9 ng/g.

Uptake and Translocation

Andersson in 1967 measured the uptake of soil applied $Hg^{203}Cl_2$ at the levels 0, 1 and 5 mg Hg/pot, and results indicated that very small quantities had been adsorbed. Barley had the highest mercury content 1.4 ng/g, thereafter oats, then clover and last Timothy grass which did not contain provable quantities. The sequence tallied with the order in which the plants sprouted in the spring. Findenegg and Haunold (3) measured the uptake by wheat straw and grain, of Hg^{203} applied at the rates of .05, 1 and 10 μg Hg/g to soils of varying textures. Results indicate that mercury is taken up by the straw in small amounts and there is little translocation to the grain even at the higher mercury treatment. Uptake was greatest in the light textured soil. Pickard, Martin and Grainger (12) have shown that potatoes grown in soil treated with mercuric oxide or mercurous chloride at 3.3 lb Hg/acre a few days before sowing gave rise to a negligible amount of mercury residue in the tubers at harvest. However, the roots were found to contain appreciable amounts of mercury.

In 1968 Yamada measured the uptake of phenyl mercuric acetate and distribution of mercury in the rice plant. Soil cultured plants adsorbed only 1% of the added P.M.A. (applied to the soil at the 13.7 μg Hg/g level). The highest mercury levels were recorded in the roots (31.4 μg Hg/g), with some translocation to the grain. Furutani and Osajima (4) have reported background mercury levels in rice much higher than in wheat. It may be that these two crops differ significantly in their ability to take up and translocate mercury or the difference could be related to the greater mobility of Hg ions in an aquatic environment.

John (1972) measured the uptake of mercury applied to a silt-loam soil at the 0, 4 and 20 μg Hg/g as mercuric chloride, by seven vegetable species and oats. At the same treatment rate and among the edible plant parts, pea seeds and oat grains accumulated the least mercury. Spinach leaves and radish tubers accumulated the highest concentrations, averaging 0.695 and 0.663 μg Hg/g of plant material respectively. There was no significant difference in the mercury content of carrot tubers grown on the three treatments. Martin (1963) found that carrots grown in soil treated with mercuric chloride contained up to 0.05 μg Hg/g when seed was sown immediately after soil treatment. An unspecified delay in seeding avoided contamination. Kosta *et al*. (1972) working in a closed ecosystem with highly increased mercury levels in the biosphere, particularly the soil, "were able to demonstrate definitely" that carrot had the ability for concentrating this element from soil containing cinnibar (HgS). Vir and Bajaj (16) immersed seeds of wheat, oats, barley and maize in a solution of radioactive mercuric chloride, and after 2 - 3 weeks growth, they found appreciable uptake and translocation of mercury throughout the seedlings. Also Saha *et al*. (1971) reported that mercury compounds, applied to the soil can be absorbed by wheat plants and translocated into the ears of grain. Methyl mercury dicyanamide was applied to soil at the 0, 3.3 and 6.6 μg Hg/g level and mercury levels in the grain from the three treatments were 10 - 14, 106 - 116 and 200 - 260 ng/g respectively in the treated plots.

Because of these rather conflicting reports regarding the uptake and translocation of mercury by plants, both field trial and growth chamber experiments were set up to measure the uptake of various forms of applied mercury and the uptake of applied mercury by different crop species.

Uptake of Different Mercury Compounds by Alfalfa

A growth chamber experiment was set up primarily to measure the uptake of different mercury carriers, added to the soil at the 10 μg Hg/g soil level as mercury, by alfalfa foliage and roots. Asquith soil was air-dried, ground to pass a 2 mm sieve and mercury added as the following carriers.

Phenyl Mercuric Acetate (PMA)

Mercuric Acetate (MA)

Mercuric Chloride ($HgCl_2$)

Mercuric Sulphate ($HgSO_4$)

Mercuric Fungicide - the active ingredients of which were PMA (7.2%) and HgCl (1.0%). The mercury equivalent was 5.0%.

Each treatment was replicated five times, and in four of each alfalfa var. Beaver was seeded to give six plants per pot. A set of control pots were also seeded.

Plant Samples

The alfalfa was harvested 67 days after seeding, and the second and third cuts were taken at 103 and 138 days respectively. After air-drying the samples were weighed and analyzed for mercury content (Tables 4 and 5).

Plant samples from the first cut contained 0.22 to 0.38 μg Hg/g D.M. No significant difference was noted between the mercury carriers, although the mercury level was four to five times the level in the control (0.7 μg Hg/g). The level in the second cut was somewhat lower at 0.15 to 0.26 μg Hg/g, compared to 0.5 μg Hg/g in the control. A similar trend can be seen in the results for the third cut.

Table 4

Dry matter yield of 3 successive cuts of alfalfa grown in mercury treated soils (10 μg Hg/g soil).

Mercury added as	Dry Matter Yield		
	1st cut	2nd cut	3rd cut
	(g/pot*)		
P.M.A.	6.30	6.95	8.22
M.A.	7.79	7.81	7.93
$HgCl_2$	6.08	6.49	7.22
$HgSO_4$	6.99	7.12	7.57
5% Hg fungicide	5.26	6.17	7.52
Control	6.40	6.77	8.06

* Results shown are the mean of 4 replicates

Table 5

Mercury content of 3 successive cuts of alfalfa grown in mercury treated soils (10 μg Hg/g soil)

Mercury added as	Mercury Content		
	1st cut	2nd cut	3rd cut
	(μg Hg/g D.M.*)		
P.M.A.	0.31	0.18	0.07
M.A.	0.27	0.26	0.10
$HgCl_2$	0.31	0.20	0.08
$HgSO_4$	0.22	0.15	0.10
5% Hg fungicide	0.38	0.24	0.11
Control	0.07	0.05	0.01

* Results shown are the mean of 4 replicates.

Root Samples

After the third harvest, a root washing with water was carried out on one replicate from each mercury treatment. After air-drying the main roots (primary, secondary and tertiary roots) and fine roots (remainder) were analyzed separately. Mercury analysis of the main roots (Table 6) showed them to contain high levels of mercury - up to 7.0 μg Hg/g D.M. Much higher levels, from 42.1 to 132.7 μg Hg/g D.M. were recorded from the analysis of the fine roots. No distinction can be made between mercury actually held within the root or on the root surface, and the possibility exists that a lot of this mercury may be held on the cation exchange at the root surface.

Table 6

Dry matter yield and mercury content of alfalfa roots grown in mercury treated soils (10 μg Hg/g soil).

Mercury added as	Dry matter yield		Mercury content	
	Main roots	Fine roots	Main roots	Fine roots
	g/pot		μg Hg/g D.M.	
P.M.A.	4.89	2.23	2.45	42.08
M.A.	4.78	2.41	7.04	56.35
$HgCl_2$	4.25	2.36	5.81	105.10
$HgSO_4$	3.60	2.11	5.99	132.70
5% Hg fungicide	4.57	2.01	3.45	44.85

Soil Samples

Mercury recovery data was obtained by analyzing the soil from two replicates of each treatment after the third harvest. Recoveries were also measured from the pots to which mercury was added but in which there was no plant growth. From Table 7 it can be seen that recoveries were good and are close to those expected from the volatility of the various mercury carriers. Lowest recoveries were obtained where mercuric chloride was the mercury carrier. This was the most volatile of the Hg containing compounds used. Recoveries were highest from the pots containing mercuric sulphate - the least volatile mercury carrier used. For the various mercury carriers used, except the 5% mercury fungicide, recoveries were highest from the pots which were left unseeded.

Table 7

Percentage recovery of mercury from mercury treated soils (10 μg Hg/g soil).

Pot #	Mercury added as	% recovery
1	P.M.A.	80.4
4		82.8
5		92.0
9	M.A.	87.0
10		85.5
11		101.0
13	$HgCl_2$	68.0
16		70.3
17		84.0
21	$HgSO_4$	95.0
22		92.0
23		100.0
26	5% Hg fungicide	89.5
27		86.5
29		87.0

Uptake of Applied Mercury by Different Plant Species

In the spring of 1972 a field trial was set up at the Canada Department of Agriculture irrigation site on the University farm. The soil of the area is a Dark Brown Chernozem belonging to the Asquith Association. The main purpose of the experiment was to study the relative uptake of a mercury fungicide incorporated into the topsoil at the rate of 10 μg Hg/g with respect to the top 0-15 cm soil by a cereal, forage, root and oil seed crop. The fungicide used was "Co-op" 5% mercury seed-dressing, normally used in the control of seed borne diseases of cereals. Mercury was present in the fungicide as Phenyl Mercuric Acetate 7.2% and Ethyl Mercuric chloride 1.0%. The mercury equivalent was 5.0%.

The plots were laid out as shown in Figure 1, and the four crops seeded were wheat var. Manitou, rape var. Span, alfalfa var. Beaver and rutabagas var. Laurentian. Mercury analysis of rape and wheat plants taken 30 days after seeding, indicated that these plants contained significant amounts of mercury. The mean mercury content of the rape plants (all results are expressed on a plant dry-matter basis), which were at the preshooting stage was 3.11 μg/g. Wheat samples at the 3-4 leaf stage contained on average 4.31 μg Hg/g.

Further plant samples of the four crops were taken for analyses at 46 and 57 days after seeding, and final samples when the crops were harvested. Results of mercury analysis are given in Tables 8, 9, 10 and 11. As growth proceeded the mercury content of each of the four

Table 8

Variation in mercury content of rutabagas var. Laurentian in μg Hg/g dry matter with time.

Days after seeding		Mercury treated plots* Rep. I	Rep. II	Rep. III	Rep. IV	Mean	Untreated plot
		(μg Hg/g D.M.)					
46		1.04	1.22	1.01	1.10	1.09	0.29
57	tops	1.01	0.97	0.68	0.85	0.88	0.15
	roots	1.14	0.55	0.69	0.96	0.84	0.10
100 (Harvest)	tops	0.55	0.43	0.49	0.54	0.50	0.08
	roots	0.17	0.10	0.10	0.13	0.13	0.06

* 5% mercury fungicide at 10 μg Hg/g soil. Plots seeded 4 days after mercury treatment.

FIGURE 1. PLAN OF FIELD EXPERIMENT*

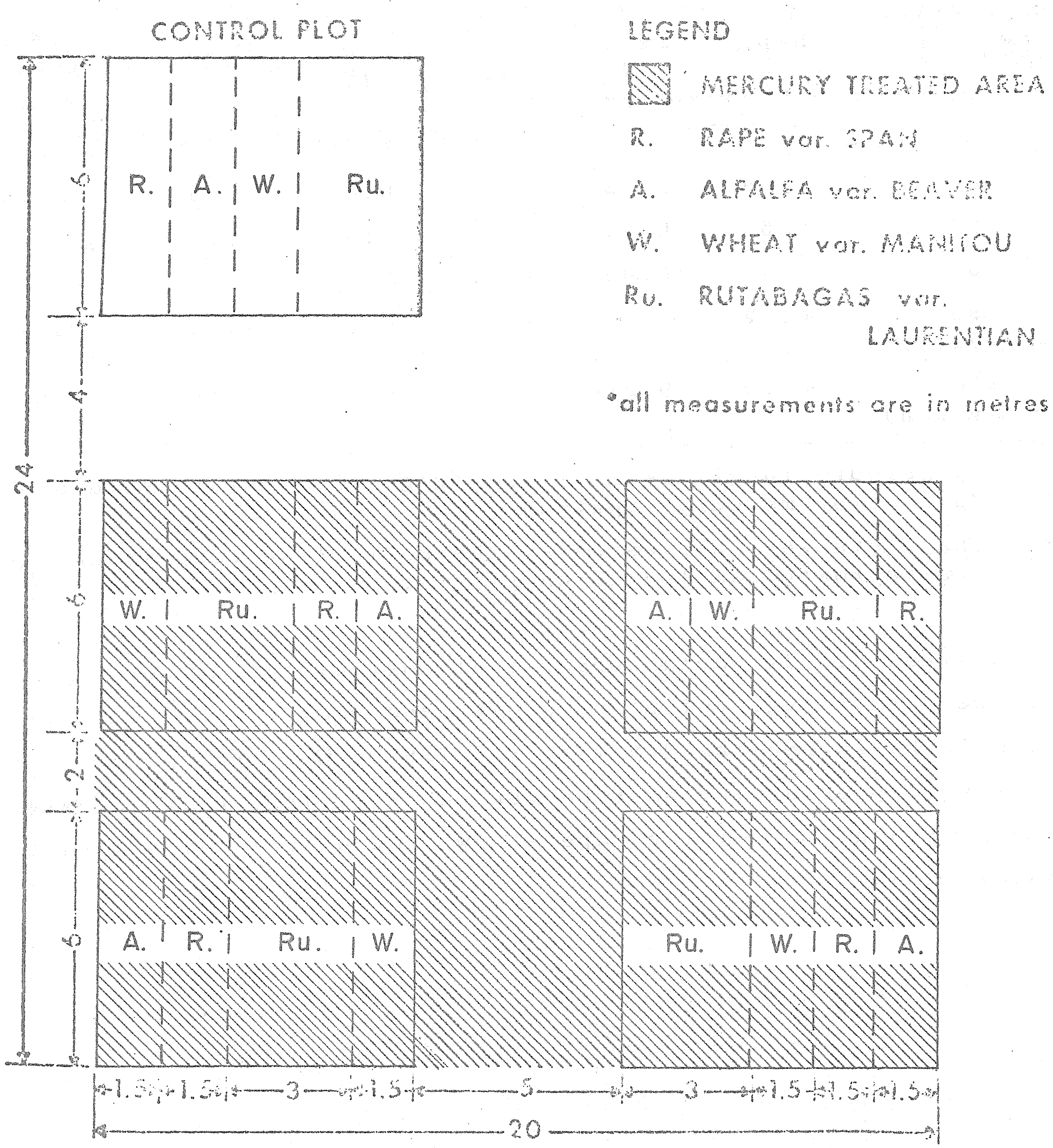

Table 9

Variation in mercury content of alfalfa var. Beaver in

µg Hg/g dry matter with time.

Days after seeding	Mercury treated plots*					Untreated plot
	Rep. I	Rep. II	Rep. III	Rep. IV	Mean	
	(µg Hg/g D.M.)					
46	1.08	1.12	0.88	0.83	0.98	0.15
57	0.57	0.83	0.55	0.50	0.61	0.12
67	0.40	0.45	0.47	0.45	0.44	0.08
88 (Harvest)	0.24	0.25	0.26	0.23	0.25	0.07
130** (Regrowth)	0.16	0.21	0.17	0.19	0.18	0.04

* 5% mercury fungicide at 10 µg Hg/g soil. Plots seeded 4 days after mercury treatment.

** The plots were harvested at the early flowering stage, and the regrowth sampled 42 days later.

crops decreased. It would appear that most of the mercury is taken up in the early stages of plant growth and what mercury in the plant is diluted by increasing dry matter weight. Only small amounts of mercury were present in the rape seed at the final harvest, whereas the mercury analysis of the rape straw showed over twice as much as wheat straw.

Table 10

Variation in mercury content of wheat var. Manitou in μg Hg/g dry matter with time.

Days after seeding		Mercury treated plots*					Untreated plot
		Rep. I	Rep. II	Rep. III	Rep. IV	Mean	
		(μg Hg/g D.M.)					
30		4.85	3.24	4.85	4.30	4.31	0.86
42		1.08	1.08	1.42	1.48	1.27	0.31
46		0.90	0.72	0.94	0.97	0.88	0.27
57		0.67	0.41	0.67	0.66	0.60	0.14
107 (Harvest)	straw	0.21	0.14	0.17	0.19	0.18	0.04
	grain	0.04	0.03	0.05	0.02	0.04	<.01

* 5% mercury fungicide at 10 μg Hg/g soil. Plots seeded 4 days after mercury treatment.

Table 11

Variation in mercury content of rape var. Span in μg Hg/g dry matter with time.

Days after seeding		Mercury treated plots* Rep. I	Rep. II	Rep. III	Rep. IV	Mean	Untreated plot
		(μg Hg/g D.M.)					
30		3.26	2.96	3.10	3.10	3.11	0.54
42		0.73	0.82	0.85	0.70	0.78	0.36
46		0.59	0.71	0.59	0.63	0.63	0.29
57		0.49	0.54	0.41	0.38	0.46	0.12
107 (Harvest)	straw	0.38	0.45	0.40	0.38	0.40	0.04
	seed	0.02	0.02	0.01	0.01	0.015	N.D.**

* 5% mercury fungicide at 10 μg Hg/g soil. Plots seeded 4 days after mercury treatment.

** N.D. - No detectable amount of mercury in this sample.

In early August of the same year, a second field experiment, using rape Var. Span as the test crop , was designed to determine the extent of immobilization of mercury, which had been applied to the soil 90 days previous to seeding. As in the first field trial, four plots were seeded and a control plot. A fixed number of plant samples were taken at 16, 20, 24, 29, 34 and 40 days after seeding, and results of the mercury analyses are given in Tables 12 and 13. The results in Table 12 show a similar trend to those observed with the rape plants in the previous field trial, i.e. with increasing dry matter weight of the plant, the mercury content decreases, except for the mercury analysis on plants harvested 29 days after seeding where there is a slight increase (Figure 2). It is interesting to note that 25 days after seeding the temperature fell below 32°F.

Figure 2

VARIATION IN THE MERCURY CONTENT OF RAPE, SEEDED IN THE FALL, WITH TIME

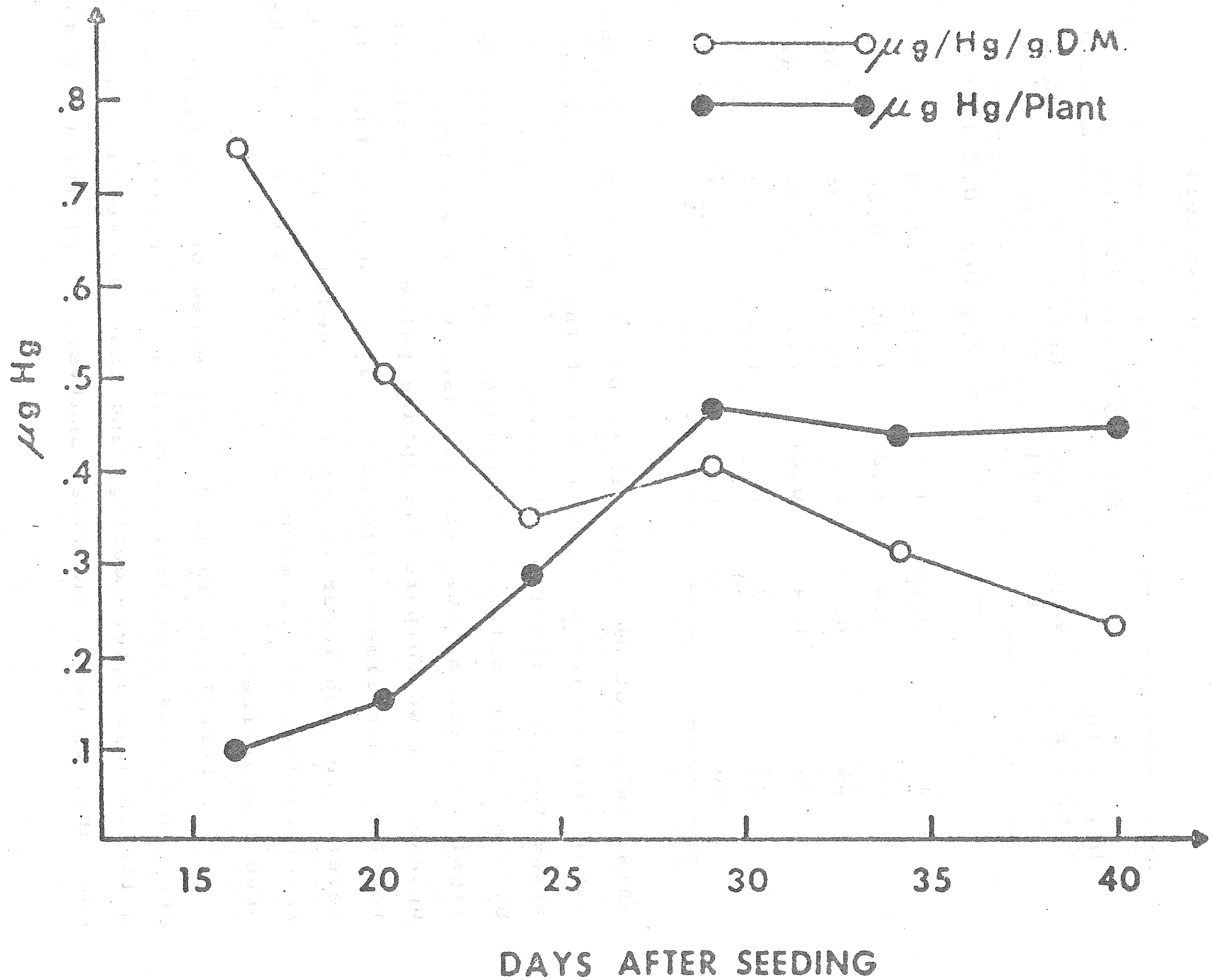

Table 12

Variation in the mercury content ($\mu g/g$ D.M.) of rape var. Span with time, grown in soil 90 days after the 5% mercury fungicide was mixed at the 10 μg Hg/g level in the top 0-15 cms soil.

Days after seeding	Mercury treated plots					Untreated plot
	Rep. I	Rep. II	Rep. III	Rep. IV	Mean	
	(μg Hg/g D.M.)					
16	0.59	0.73	0.62	1.02	0.74	0.06
20	0.27	0.62	0.49	0.60	0.50	0.06
24	0.23	0.39	0.38	0.38	0.35	0.03
29	0.41	0.41	0.41	0.39	0.41	0.02
34	0.34	0.31	0.31	0.32	0.32	0.02
40	0.16	0.23	0.30	0.27	0.24	0.02

This would probably have killed off a small proportion of the bacterial population in the surface layer of the soil. Andersson (1) has suggested that fixation of mercury is probably due to a great extent by a microbial nature. Therefore, one can conclude that when the frost occurred mercury was released which was then available for plant uptake. If we compare the uptake patterns with the rape seeded in the first experiment (Figure 3), it is obvious that the mercury levels are much lower in the rape seeded in plots 90 days after incorporation of the mercury fungicide, i.e. most of the mercury has been immobilized.

The results in Table 13 (expressed on a μg Hg/plant basis) show that there was a gradual increase in the mercury content per plant until 29 days after seeding, and then the mercury levels remain fairly constant, up until the final analysis of plants taken 40 days after

Figure 3

VARIATION IN THE MERCURY CONTENT OF RAPE WITH TIME

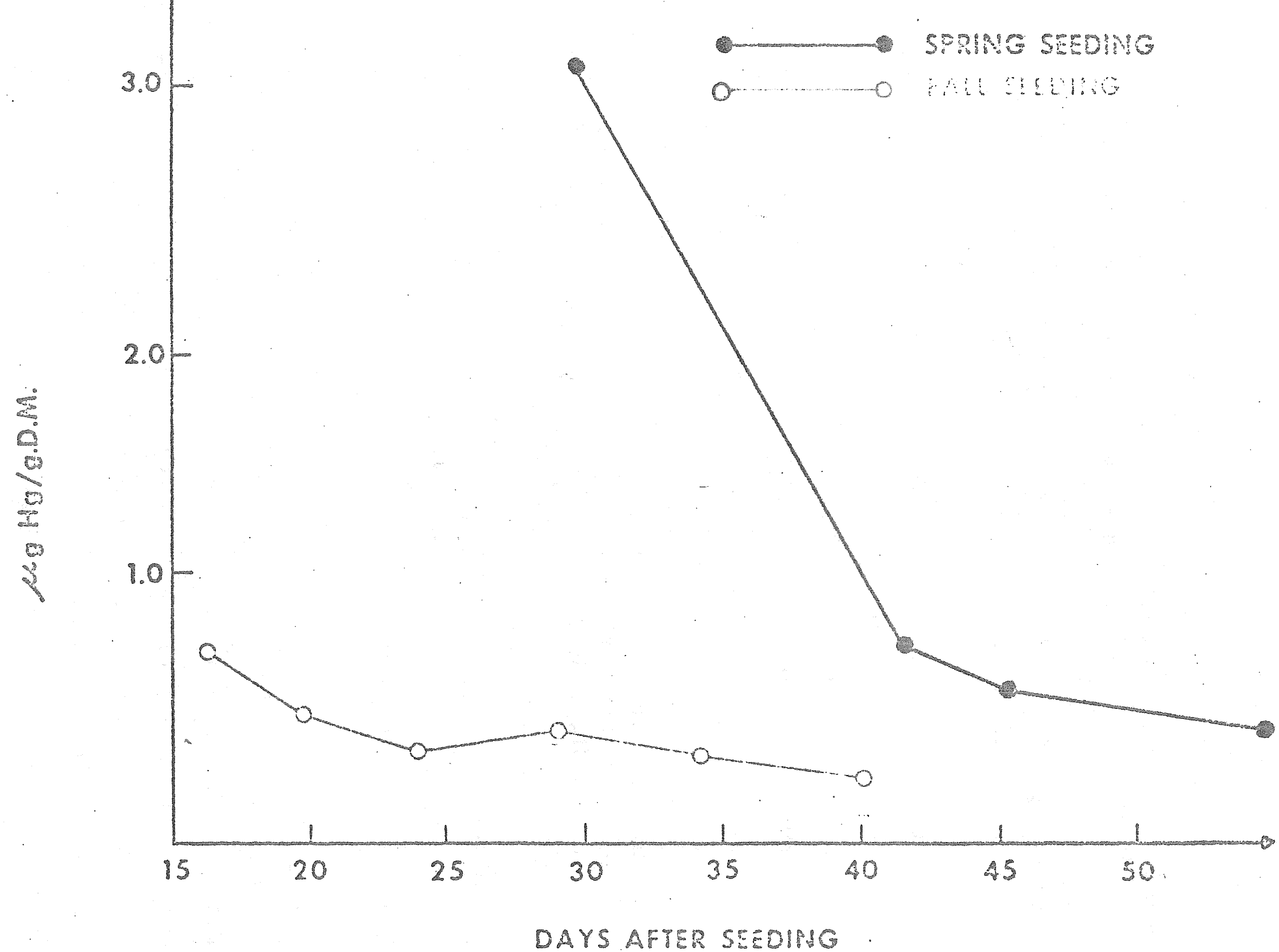

Table 13

Variation in the mercury content (μg) of rape plants* var. Span with time, grown in soil 90 days after the 5% mercury fungicide was mixed at the 10 μg Hg/g level in the top 0-15 cms soil.

Days after seeding	Mercury treated plots					Untreated plot
	Rep. I	Rep. II	Rep. III	Rep. IV	Mean	
	(μg Hg)					
16	0.08	0.12	0.08	0.14	0.11	0.01
20	0.09	0.21	0.16	0.19	0.16	0.02
24	0.19	0.32	0.33	0.35	0.30	0.04
29	0.50	0.47	0.51	0.42	0.48	0.03
34	0.47	0.42	0.46	0.44	0.45	0.03
40	0.33	0.47	0.55	0.47	0.46	0.04

* The mercury content per plant in μgs was obtained by multiplying the mean mercury content per plant in terms of μgs Hg/g dry matter by the mean dry matter weight per plant.

seeding (Figure 2). This confirms the results from the earlier experiment which indicated that mercury is mainly taken up in the early stages of plant growth.

Conclusion

Our present knowledge regarding mercury in the soil is very limited. The experiments reported here have provided much needed information on background levels of mercury in plants and soils, and have clearly indicated that more attention should be paid to the effect of the rooting system of plants, the role of the bacterial population, as well as soil clay and organic matter as causes of the immobilization, of mercury. Research is needed to determine the extent of 'labile' mercury in the soil, in contrast to the results given here which represent total mercury levels. Little research has been done on the forms of mercury present in soil or plant material.

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