Residues as high as 42.80 ppm were detected in a composite sample of nightshade, ironweed, and careless weed collected from a treated area of a dryland field 7 days following treatment, decreasing to 0.96 ppm 29 days after treatment.

Residues were also detected in untreated sections of dryland fields approximately 12–13 ft from the treated rows. Seven days following treatment, residues of 0.02 ppm were detected in a sample collected approximately 13 ft from treated rows. One sample of careless weed collected *ca.* 3 ft from treated rows showed a residue of 2.37 ppm 29 days after treatment; another sample of careless weed collected 13 ft from treated rows showed 2.85 ppm of aldicarb and/or metabolites. One thistle sample collected approximately 13 ft from the treated area showed a residue of 19.64 ppm. No detectable residues were found in untreated, noncultivated areas adjacent to the treated fields.

Table II presents residue data for samples of weeds and grass collected from treated fields receiving irrigation. Highest residues detected from treated areas were 2.09 ppm in a sample of careless weed 54 days after treatment, 2.94 ppm in a composite sample of Johnsongrass and ironweed 74 days following treatment, 1.85 ppm in a composite sample of careless weed, Johnsongrass, and Colorado grass, and 3.37 ppm in a composite sample of Johnsongrass and ironweed. Samples collected from untreated areas of the fields showed significant residues in only three samples: 0.12 ppm in a sample of nightshade collected 62 days after application; 23.59 ppm in a composite sample of careless weed and Johnsongrass collected 54 days following treatment; and 0.55 ppm in a sample of careless weed collected 51 days after treatment. All were collected approximately 13 ft from treated areas. Detectable residues were found in only three samples from adjacent untreated, noncultivated areas (0.03, 0.02, and 0.01 ppm) in material collected 51, 51, and 54 days, respectively, after treatment.

Residues were detected in only one wildlife sample, an oriole bird with 0.07 ppm of aldicarb and/or metabolites.

DISCUSSION

Aldicarb was found to be a pesticide which translocated into grasses and weeds in treated and untreated dryland and irrigated fields within a matter of weeks following treatment. No significant movement of the pesticide into uncultivated areas adjacent to either dryland or irrigated fields was noted. No indication of significant introduction into the biological food chain was noted since only one bird of the 14 sampled showed detectable residues; no detectable residues were found in any of the eight animals sampled.

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Mercury and Methylmercury Content of Agricultural Crops Grown on Soils Treated with Various Mercury Compounds

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Beans, cabbage, carrots, millet, onions, potatoes, and tomatoes were grown on silt loam, gravelly loam, and muck soils treated with 1 and 10 ppm of mercuric chloride, methylmercury dicyandiamide (PAN), or phenylmercuric acetate. Appreciable concentrations of methylmercury were present only in PAN-treated soils and in beans, millet, and tomatoes grown on those soils. Total mercury was usually less than 0.1 ppm in the edible plant portions, with the highest concentrations occurring most generally when growth occurred on the gravelly loam treated with PAN. Onion bulbs absorbed up to 1.1 ppm of total mercury. The highest concentrations of total mercury in plant stems and leaves were attained in potatoes and tomatoes.

The recently discovered ubiquitous presence and consequences of mercury in the environment have been amply reviewed (Goldwater, 1971; Peakall and Lovett, 1972; Saha, 1972). Much research has been done on various aspects of mercury in aquatic, animal, and human systems. Extensive data are available on mercury residues in plants resulting from foliar applications (Smart, 1968). Data are sparse, however, on the absorption of mercury into plants from soils.

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Mercury in soil may result from fungicide applications, air pollution, or that present natively. An analytical survey of mercury in 912 samples of soil taken throughout the United States showed levels ranging from 55 to 4600 ppb, with a geometric mean of 71 ppb (Shacklette *et al.*, 1971). From limited data available, it appears that plants rarely contain mercury concentrations above 500 ppb (Shacklette, 1970). Plants may absorb higher concentrations of mercury when grown in proximity to mercury ore deposits or mines but it is possible that mercury in the air in these regions may contribute to their total content (Byrne and Kosta, 1970). It has been reported that jagged chickweed (*Holosteum umbellatum*) and certain algae may concen-

Table I. Recovery of Mercury and Methylmercury from Control Samples

Sample	Added, ppb	Recovery, %	Sample	Added, ppb	Recovery, %
		Mercuric chloride added			
Bean pods	50	98	Millet shoots	50	70
	100	104		100	63
	300	92		300	75
Bean shoots	50	110	Onion bulbs	50	94
	100	105		100	97
	300	112		300	97
Cabbage heads	50	86	Onion tops	50	88
	100	99		100	93
	300	94		300	95
Carrot roots	50	70	Potato tubers	50	84
	100	73		100	82
	300	92		300	94
Carrot tops	50	84	Potato shoots	50	76
	100	92		100	81
	300	95		300	68
Millet grain	100	84	Tomato fruit	50	72
	200	85, 87		300	83
	600	97	Howard gravelly loam soil	1000	74, 123, 93, 86, 88, 94, 87
Eel silt loam soil	1000	101, 94, 82, 77, 86, 100, 106			
Oswego muck soil	1000	105, 98, 157, 79, 97, 116, 68			
		Methylmercuric chloride added			
Bean pods	8.7	79, 85	Potato tubers	8.7	77
Bean shoots	8.7	83	Potato shoots	8.7	81
Cabbage heads	8.7	98	Tomato stalks	8.7	64
Carrot roots	8.7	77	Eel silt loam soil	8.7	89
Carrot tops	8.7	81	Howard gravelly loam soil	8.7	90
Millet grain	8.7	81, 64	Oswego muck soil	8.7	83
Onion bulb	8.7	65			

trate mercury, but no quantitative data are available (Rankama and Sahama, 1950).

Limited published information is available on mercury in crops resulting from agricultural applications of mercury compounds to soil. Negligible residues (below 0.05 ppm) of mercury in potato tubers were reported when the plants were grown on soils treated with phenylmercuric chloride (Smart, 1964), mercuric oxide, or mercurous chloride and analyzed by spectrophotometric determination of mercuric dithizonate (Pickard *et al.*, 1962). These latter workers also found negligible residues of total mercury in potato tubers grown on soil treated with various inorganic mercury compounds. Using flameless atomic absorption analysis, John (1972) found detectable levels of mercury (greater than about 0.01 ppm) in various vegetables and oat grain grown on soil treated with mercuric chloride at rates up to 20 ppm. A small but significant concentration of mercury resulted in wheat grain grown on soil treated with Panogen PX (methylmercury dicyandiamide) (Saha *et al.*, 1970).

The behavior of certain mercury compounds in soil has been studied. Phenylmercuric acetate has been shown to decompose in soil with the production of diphenylmercury and other unidentified metabolites (Matsumura *et al.*, 1971). Phenyl-, ethyl-, and methylmercury compounds in soil all partially decomposed with production of mercury vapor (Kimura and Miller, 1964). They found that about 14 to 16% of the phenylmercuric acetate in soil was lost as volatile mercury vapor. Sixty to 70% remained intact in the soil, with about 20% remaining uncharacterized. Methylmercury dicyandiamide was converted (up to 14%) to methylmercury, which vaporized out of the soil. Only a small amount of mercury vapor was produced. Mercury has been shown to be methylated by microorganisms in aquatic bottom muds to methylmercury and dimethylmercury (Jensen and Jernelöv, 1969; Wood *et al.*, 1968). Dimethylmercury is water-insoluble and very volatile and would probably be rapidly lost. Methylmercury is water-soluble and is extremely toxic when ingested, since it is believed to penetrate the blood–brain barrier and to cause irreversible brain damage in mammals. Mercuric ion may be strongly adsorbed in soils on the surfaces of clays, precipitates, and organic matter. Calcium or sodium chloride, as used for deicing roads, may run off and release mercuric ion from contaminated freshwater sediments (Feick *et al.*, 1972).

In the work reported here, a study was made of the extent of absorption by plants of mercury from soils treated with various mercury compounds. The possibility of the methylation of mercury in the soil–plant system was also investigated.

EXPERIMENTAL SECTION

Greenhouse Studies. Three soils were used: Eel silt loam and Howard gravelly loam from Freeville, N. Y., and Oswego muck from Fulton, N. Y. The soils were freshly dug from fields in the areas. The soils were mixed and separate portions were treated with 0 (control), 1, or 10 ppm of one of the compounds: mercuric chloride (MC), phenylmercuric acetate (PMA), or Panogen (PAN) (methylmercury dicyandiamide). The soil was rotated in a concrete mixer, while 250 ml of MC or PAN (formulated as Pan-O-Drench) in water or PMA in absolute ethanol was applied using an Agway No. 113 Squire Applegate 1.5-gal hand sprayer. Each liter of aqueous MC was acidified and partitioned six times with successive 100-ml portions of benzene before use to remove methylmercury impurities (Westoo, 1967). The rate of application was

equivalent ppm by weight of the compound, expressed as mercury per weight of soil at field capacity. Field capacity of the soils were: silt loam, 30%; gravelly loam, 25%; and muck, 95%.

The crops studied were: bush bean (*Phaseolus vulgaris*), cultivar Tendergreen; cabbage (*Brassica oleracea* var. *capitata*); cultivar Golden Acre; carrot (*Daucus carota* var. *sativa*), cultivar Scarlet Nantes; Japanese millet (*Echinochloa crusgalli* var. *frumentacea*); onion (*Allium cepa*), cultivar Downing Yellow; potato (*Solanum tuberosum*), cultivars Katahdin and Sebago; and tomato (*Lycopersicon esculentum*) cultivar New Yorker. Within 4 to 24 hr after soil application of mercury compounds, all crops were seeded in pots, 9 in. in diameter, except for potatoes, for which 12-in. pots were used. In pots where plants did not emerge, reseeding was carried out within 12 days of the initial planting. The weight in pounds of soils used, respectively, in the 9- and 12-in. pots were 13 and 30 for the silt loam, 16 and 39 for the gravelly loam, and 8 and 18 for the muck. The number of plants grown to maturity in each pot were: bean, 2; cabbage, 1; carrot, 3; millet, 5; onion, 3; potato, 1; and tomato, 1. All treatments were replicated three times. The greenhouse experiment therefore involved a factorial of 7 crops × 3 soils × 3 mercury compounds × 3 application rates × 3 replicates. All plants were fertilized weekly and watered daily. Care was taken to avoid splashing soil on the aerial portions of plants. The plants were harvested and sectioned into parts. Plant roots were washed exhaustively to remove adhering soil particles. Soils were dried, mixed, and sampled. All samples were placed in polyethylene bags and frozen prior to analysis.

Mercury Analysis. The three replicates of plants and soils in each treatment were combined. The various plant parts were subdivided by homogenizing in a blender or chopping in a food cutter. One gram of the well-mixed sample was dried and combusted in an oxygen flask, as previously described (Gutenmann and Lisk, 1960). Soils were dried and mixed, and 0.5 g of the soil was similarly combusted (White and Lisk, 1970). Analysis of total mercury was performed by flameless atomic absorption analysis (Hatch and Ott, 1968). Analysis of samples for methylmercury was performed by the procedure of Bache and Lisk (1971) using gas chromatography with the microwave-powered plasma emission detector. The methods were sensitive to about 5 ppb of total mercury and 1 ppb of methylmercury.

RESULTS AND DISCUSSION

The recoveries of total mercury added as mercuric chloride and methylmercury added as methylmercuric chloride to plants and soils are listed in Table I. The recoveries of mercuric chloride from soils were probably more variable than in the case of plants, since the level of fortification in soils (1 ppm) was higher than that for plants (0.05 to 0.6 ppm). Since the flameless atomic absorption method for mercury is highly sensitive, a greater dilution error would have been manifested in the case of soil analysis. The recoveries of methylmercuric chloride were expectedly less than quantitative. The method of analysis of Bache and Lisk (1971) included essentially the isolation procedure used by Westoo (1966, 1967, 1968), who found that recovery of methylmercury was typically about 70% owing to an approximate 30% loss of methylmercury because of unfavorable solvent partition coefficients.

Residues of total mercury and methylmercury in plants and methylmercury in soils (after plant growth) are given in Tables II and III. Several observations are apparent from the data. The highest concentrations of total mercury were most generally attained in plants grown on Howard gravelly loam and when treated with PAN. The total concentration of mercury consistently reached the highest concentrations in plant stems and leaves in the case of potatoes (1045 ppb) and tomatoes (341 ppb). Onion bulbs absorbed up to 1087 ppb of mercury in the Howard gravelly loam. The nonedible aerial portions of carrots (231 ppb), millet (91 ppb), potatoes (1045 ppb), and tomatoes (231 ppb) showed comparatively high levels of total mercury on the Howard gravelly loam soil treated with MC, especially at the 10-ppm rate of application. Based on the normal levels of mercury found in soils (Shacklette *et al.*, 1971), the rates of addition of mercury compounds used in this investigation are admittedly high. Since the plants grown on these soils are still comparatively low in total mercury, it may indicate that plants generally do tend to exclude mercury during absorption depending on the tenacity of mercury fixation in the soil.

Methylmercury analysis was performed only on those plants showing the most elevated total mercury concentrations. Residues of methylmercury were present in appreciable concentrations only in plants and soils in the case of the PAN treatments. The residues were highest in beans (125 ppb) and potatoes (183 ppb).

It is interesting to speculate concerning the sequence of reactions leading to methylmercury residues in the soils and plants. Kimura and Miller (1964) reported that most of the PMA and PAN remained intact in soil over a period of 35 days. Conversely, Spanis *et al.* (1962) reported that PAN was inactivated by soil microorganisms. In this study, within experimental error, analysis of total mercury in all soils indicated little loss of it during the growing period, regardless of the compound applied or the rate of application. In the PAN-treated soils only a relatively small portion of the total mercury was present as methylmercury in the soil. The initial strong hydrochloric acid overnight extraction of soil for methylmercury would convert any remaining intact PAN to methylmercuric chloride. (Recovery of 0.6 ppm of PAN added to soil was 91% using the hydrochloric acid extraction, followed by determination of methylmercury as the chloride (Bache and Lisk, 1971).) The relatively low concentrations of methylmercury found in the PAN-treated soils would thus indicate that most of the PAN has been degraded in the soil to mercuric ion or some form other than intact methylmercury. If this is correct, the plants then absorbed intact methylmercury from the soil. Since barely detectable residues of methylmercury were found in plants grown on MC-treated soils, it is not plausible to hypothesize that the plants methylated mercuric ion following its uptake. Since this study indicates that plants apparently can absorb intact methylmercury from soils, it would be interesting to analyze rice from various sources for methylmercury, since rice grows on submerged soils in which methylation of native soil mercury may occur.

Several observations are noteworthy concerning plant growth. Root and shoot development was excellent in the muck, satisfactory in the silt loam, and poor in the gravelly loam. Phytotoxicity was absent in muck, intermediate in the silt loam, and greatest in the gravelly loam. In the silt loam treated with 10 ppm of any of the mercury compounds, emergence of all plants (except potato, which was unaffected) was delayed 5 to 6 days, as compared to the controls. After reaching maturity, however, there was no discernible difference between any of the treated plants as compared to the controls. In the gravelly loam nearly all of the potted soils receiving 10 ppm of the various mercury compounds had to be reseeded two to three times to effect germination. About half of those at the 1-ppm level had to be reseeded. Considerable soil compaction was evident in the gravelly loam within 2 weeks after planting and drainage was poor. Therefore, plant roots generally grew into only the top 1–2 in. of soil.

In conclusion, it should be mentioned that one must consider the analytical methods employed when comparing the results of this investigation with those of previous researchers. Before the advent of oxygen flask combustion

Table II. Residues of Total Mercury and Methylmercury in Plants

Soil	Mercury compound applied	Application rate, ppm as mercury	Residue[a] (ppb (fresh weight) as mercury and corrected for control) in Edible vegetables[b] Total mercury	Edible vegetables[b] Methylmercury	Stems and leaves Total mercury	Stems and leaves Methylmercury
			BEANS			
Eel silt loam	MC	0	nd[c]		37	
		1	nd		nd	
		10	nd		nd	
	PAN	0	5		19	
		1	22		28	
		10	21		38	
	PMA	0	nd		34	
		1	nd		nd	
		10	nd		nd	
Howard gravelly loam	MC	0	6		57	nd
		1	nd		1	nd
		10	6		33	nd
	PAN	0	24	2	83	2
		1	37	48	52	46
		10	181	125	90	28
	PMA	0	6		48	
		1	nd		nd	
		10	nd		nd	
Oswego muck	MC	0	7		37	
		1	nd		nd	
		10	5		5	
	PAN	0	7		45	
		1	14		nd	
		10	59		15	
	PMA	0	12	nd[d]	32	
		1	nd	nd	3	
		10	156	nd	9	
			CABBAGE			
Eel silt loam	MC	0	53			
		1	10			
		10	nd			
	PAN	0	55			
		1	22			
		10	32			
	PMA	0	25			
		1	5			
		10	nd			
Howard gravelly loam	MC	0	12	1		
		1	10	nd		
		10	43	nd		
	PAN	0	29	1		
		1	nd			
		10	133	3		
	PMA	0	22			
		1	4			
		10	nd			
Oswego muck	MC	0	16			
		1	39			
		10	7			
	PAN	0	33			
		1	nd			
		10	2			
	PMA	0	16	1		
		1	1	nd		
		10	31	nd		
			CARROTS			
Eel silt loam	MC	0	14	nd	34	
		1	14	nd	nd	
		10	12	nd	4	
	PAN	0	16		25	
		1	9		6	
		10	15		5	

Table II *(Continued)*

Soil	Mercury compound applied	Application rate, ppm as mercury	Residue[a] (ppb (fresh weight) as mercury and corrected for control) in Edible vegetables[b] Total mercury	Edible vegetables[b] Methylmercury	Stems and leaves Total mercury	Stems and leaves Methylmercury
	PMA	0	11		26	
		1	nd		nd	
		10	18		13	
Howard gravelly loam	MC	0	36	nd	39	nd
		1	nd		nd	
		10	73	nd	231	nd
	PAN	0	6	0	26	nd
		1	31		104	
		10	279	3	214	1
	PMA	0	36	nd	41	nd
		1	nd	nd	nd	nd
		10	29	nd	17	nd
Oswego muck	MC	0	15		25	
		1	nd		1	
		10	nd		20	
	PAN	0	17		32	
		1	3		5	
		10	23		16	
	PMA	0	23		32	
		1	nd		nd	
		10	10		9	
			MILLET			
Eel silt loam	MC	0	30		17	
		1	nd		1	
		10	nd		1	
	PAN	0	32	nd	24	
		1	22	19	nd	
		10	8	9	2	
	PMA	0	30		32	
		1	2		nd	
		10	nd		nd	
Howard gravelly loam	MC	0	50	1	16	nd
		1	22	2	8	nd
		10	64	1	91	nd
	PAN	0	48	nd		
		1	94	81	44	
		10	58	20	123	
	PMA	0	36		20	nd
		1	60		5	2
		10	2		27	nd
Oswego muck	MC	0	12		16	
		1	nd		7	
		10	6		9	
	PAN	0	12	nd	20	
		1	18	16	5	
		10	40	28	6	
	PMA	0	22		28	
		1	nd		nd	
		10	nd		nd	
			ONIONS			
Eel silt loam	MC	0	nd		24	
		1	3		nd	
		10	5		4	
	PAN	0	13		23	
		1	3		4	
		10	9		1	
	PMA	0	5	2	17	
		1	4	nd	8	
		10	34	nd	nd	
Howard gravelly loam	MC	0	6	nd	8	
		1	10		nd	
		10	1087	3	19	

Table II *(Continued)*

Soil	Mercury compound applied	Application rate, ppm as mercury	Residue[a] (ppb (fresh weight) as mercury and corrected for control) in Edible vegetables[b] Total mercury	Edible vegetables[b] Methylmercury	Stems and leaves Total mercury	Stems and leaves Methylmercury
	PAN	0	21	1	22	
		1	nd		nd	
		10	1044	6	13	
	PMA	0	13	nd	13	
		1	5		nd	
		10	113	nd	13	
Oswego muck	MC	0	10		18	
		1	nd		nd	
		10	nd		5	
	PAN	0	7		11	
		1	16		nd	
		10	33		28	
	PMA	0	nd		20	
		1	nd		16	
		10	10		8	
			POTATOES			
Eel silt loam	MC	0	7		71	
		1	2		nd	
		10	6		nd	
	PAN	0	5		62	1
		1	83		86	27
		10	53		58	17
	PMA	0	5		101	
		1	2		nd	
		10	3		nd	
Howard gravelly loam	MC	0	11	nd	50	nd
		1	nd	nd	31	nd
		10	130	nd	1045	nd
	PAN	0	8	1	67	nd
		1	44	56	nd	6
		10	327	80	503	9
	PMA	0	5		28	nd
		1	6		38	1
		10	16		69	1
Oswego muck	MC	0	9		92	
		1	nd		nd	
		10	nd		nd	
	PAN	0	5	nd	60	
		1	67	65	14	
		10	196	183	42	
	PMA	0	nd		68	
		1	5		3	
		10	18		15	
			TOMATOES			
Eel silt loam	MC	0	18		76	nd
		1	23		42	
		10	13		68	nd
	PAN	0	43		109	
		1	nd		nd	
		10	nd		8	
	PMA	0	7		86	nd
		1	nd		43	nd
		10	11		9	nd
Howard gravelly loam	MC	0	nd		86	nd
		1	7		nd	
		10	13		231	nd
	PAN	0	nd		91	nd
		1	nd		nd	2
		10	13		341	3
	PMA	0	nd		95	
		1	nd		nd	
		10	nd		nd	

Table II *(Continued)*

Soil	Mercury compound applied	Application rate, ppm as mercury	Residue[a] (ppb (fresh weight) as mercury and corrected for control) in Edible vegetables[b] Total mercury	Edible vegetables[b] Methylmercury	Stems and leaves Total mercury	Stems and leaves Methylmercury
Oswego muck	MC	0	52		78	
		1	nd		4	
		10	nd		nd	
	PAN	0	32		122	
		1	nd		nd	
		10	nd		nd	
	PMA	0	14		164	
		1	nd		nd	
		10	nd		nd	

[a] Residue not corrected for percent. [b] Except millet grain recovery. [c] Not detectable; *i.e.*, less than 5 ppb. [d] Not detectable; *i.e.*, less than 1 ppb.

Table III. Residues of Methylmercury in Soils after Plant Growth

Soil	Mercury compound applied	Application rate, ppm as mercury	Residue (ppb fresh weight) as mercury and corrected for control
Eel silt loam	MC	0	0
		1	6
		10	4
	PAN	0	0
		1	107
		10	333
	PMA	0	0
		1	0
		10	0
Howard gravelly loam	MC	0	0
		1	1
		10	1
	PAN	0	0
		1	59
		10	367
	PMA	0	0
		1	4
		10	4
Oswego muck	MC	0	4
		1	16
		10	2
	PAN	0	3
		1	707
		10	1707
	PMA	0	11
		1	8
		10	nd

and flameless atomic absorption analysis, the determination of trace levels of mercury was a formidable analytical challenge owing to vaporization losses during wet ashing, incomplete combustion, interference from other cations (such as copper), and lack of sensitivity. Unless the data were accompanied by adequate recovery studies to verify analytical accuracy, the results were often gravely in doubt. As regards methylmercury analysis, the microwave emission detector employed here is a highly selective (the selectivity ratio is over 10,000 to 1) and sensitive device for specific mercury analysis which, interfaced with gas chromatography, has been used by us for analysis of methylmercury in hundreds of various biological samples.

It should also be mentioned that an effort was made to evenly distribute the mercury compounds throughout the soil prior to potting by atomizing solutions of the compound into the mass of soil while it tumbled in a cement mixer with baffles. Even distribution will not result if instead a few milliliters of the compound are pipetted onto several pounds of soil, followed by quartering the whole mass. This leads to pockets of soil with highly concentrated mercury, which one must tacitly assume the plant roots will contact. Soil fixation and root absorption of mercury would not expectedly be comparable under these conditions. Finally, it should be pointed out that it was virtually impossible to thoroughly remove all adhering soil microparticles from the fine, fibrous plant roots. Therefore, no attempt was made to analyze plant roots in this study. It is suspected that previously reported high levels of mercury in plant roots may have been due to soil contamination.

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