Diazinon. II. Residues in plants, soil, and water

By

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Contents

I. Introduction

Diazinon [*O*,*O*-diethyl *O*-(2-isopropyl-4-methylpyrimidin-6-yl) phosphorothioate] was introduced as a plant protection product in 1954. On a variety of cultivated plants and in soil, diazinon controls chewing, mining, and nonresistant sucking pests (e.g., locusts, caterpillars, beetles, leaf miners, gall midges, maggots, fruit flies, sawflies, aphids, jassids, and bugs).

Many food and feed crops have been analyzed during the past years for residues of diazinon and its transformation products. The present objective is to review the available residue data for diazinon in plant materials, soil, and water. Each section on a group of crops contains an assessment of the quantitative residue aspects including a brief discussion of the distribution and metabolism pattern.

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F. A. Gunther et al. (eds.), *Residue Reviews*

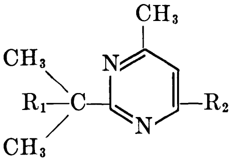
Diazinon is first degraded by hydrolysis of the thio-phosphorus ester with subsequent side-chain hydroxylation of the resulting 2-isopropyl-4-methyl-6-hydroxypyrimidine. This side-chain hydroxylation may also occur before cleavage of the ester bond as shown by the presence of small amounts of "hydroxy-diazinon." Small levels of diazoxon indicate that oxidation is very limited or that the oxon is hydrolyzed as rapidly as it is formed.

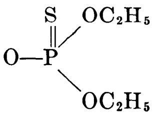
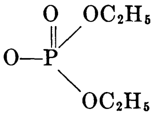
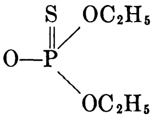
The structures and chemical names of diazinon and its degradation products mentioned in this review are as shown in Table I.

II. Diazinon in raw agricultural products

About 70 agricultural products have been evaluated for residues. The results are summarized in Tables II through VII. The data stem from field experiments carried out in different climates according to good agricultural practice, involving a set of different application

Table I. *Diazinon and its degradation products.*



Compound	R_1	R_2	Chemical name
Diazinon	H		*O,O*-diethyl *O*-(2-isopropyl-4-methylpyrimidin-6-yl) phosphorothioate
Diazoxon	H		*O,O*-diethyl *O*-(2-isopropyl-4-methyl-pyrimidin-6-yl)phosphate
"Hydroxy-diazinon"	HO		*O,O*-diethyl *O*-[2-(1′-hydroxy-1′-methyl)ethyl-4-methyl-pyrimidin-6-yl] phosphorothioate
"Pyrimidinol"	H	OH	2-isopropyl-4-methyl-6-hydroxypyrimidine
"Hydroxy-pyrimidinol"	HO	OH	2-(1′-hydroxy-1′-methyl)ethyl-4-methyl-6-hydroxypyrimidine

rates. Most of the residue data have been obtained by the Analytical Department of Geigy Agricultural Chemicals, a Division of the former *Geigy Chemical Corp.*, Ardsley, N.Y., U.S.A., and by the residue laboratory of the former J. R. GEIGY S. A., BASLE, Switzerland, now *CIBA-GEIGY* Ltd.

The half-life periods of diazinon in the various plants mentioned in this review are estimated from degradation curves (see data in Tables II to VI) and the "Diazinon Monograph" of the FAO/WHO (1971). The various analytical methods used for residue determination are indicated in the tables. For details of the methods the reader is referred to the preceding review by EBERLE (1974) in this volume.

a) *Fruits and nuts* (Table II)

The residues in pome fruits and grapes ranged from 0.5 to one p.p.m. immediately after treatment and were on the average 0.4 p.p.m. in apples and 0.15 p.p.m. in grapes after ten days. In stone fruits, e.g., plums and cherries, initial residues of two to five p.p.m. were found, which dissipated rapidly to 0.2 p.p.m. and below ten days after treatment. On rapidly growing fruits such as cherries and grapes the residues decreased rapidly; half-life periods of two and three days, respectively, were observed. In apples and peaches the half-life value was five days. Like other insecticides, diazinon dissipates more slowly on peaches, probably because of the hirsute surface of the fruits. Residue levels in nuts and berry fruits were extremely low, amounting to <0.05 p.p.m. after seven days.

1. Distribution. — According to STOBWASSER (1962), 90 percent of diazinon residues on and in apples remained in the peel and only 2.4 percent reached the pulp of the fruit. Juice and pulp of citrus fruits were completely free of residues, the peel obviously serving as an efficient barrier against deep penetration (GUNTHER *et al.* 1958).

2. Metabolites. — The only metabolite found in pome fruits was diazoxon, the corresponding oxygen analog. Maximum levels in apples and pears were 0.004 and 0.003 p.p.m., respectively, at any time after application. At harvest, the level dropped to less than 0.002 p.p.m. In cherries no diazoxon was found (EBERLE and NOVAK 1969).

b) *Vegetables* (Table III)

In addition to the usual treatment of the upper plant parts the activity of diazinon is frequently exploited by soil applications in vegetable crops.

Carrots have been examined with particular care as they are often eaten uncooked and are processed for babyfood. Furthermore, it is known that pesticides are likely to be stored in carrots due to the rather high content of oil which protects them from metabolic attack

Table II. *Residues of diazinon in fruits and nuts.*

Sample	Dosage (% a.i. or kg. a.i./ha.)	Formulation[a]	No. of applications	Days after last application	Residue (p.p.m.)[b]	Analytical method[c]	Country and year	Reference[d]
Almonds								
Nuts	0.8 kg.	WP	2	0	0.15	S	U.S.A. (1966/67)	G.A.
				8	0.09	S		
				16	<0.05	S		
Shells	0.8 kg.	WP	2	0	2.3	S		
				8	0.12	S		
				16	0.05	S		
Apples								
Fruits	0.02 %	EC	1	0	0.75	S	Germany G.F.R. (1961)	Stobwasser (1962)
				3	0.45	S		
				7	0.15	S		
				14	0.25	S		
Fruits	0.04 %	WP	3	3	1.8	GC	Switzerland (1966)	G.B.
				5	1.0	GC		
				7	0.6	GC		
				14	1.2	GC		
				28	0.3	GC		
				49	0.2	GC		
				63	0.02	GC		
Fruits	0.04 %	WP	3	0	0.4	GC	Switzerland (1966)	G.B.
				4	0.2	GC		
				6	0.2	GC		
				11	0.2	GC		
				15	0.08	GC		
				22	0.05	GC		
Fruits	0.04 %	WP	5	0	0.9	GC	Switzerland (1966)	G.B.
				2	1.0	GC		
				5	0.9	GC		
				7	0.8	GC		
				14	0.3	GC		
				21	0.4	GC		
				35	0.3	GC		
Apricots								
Fruits	0.06 %	EC	2	0	1.5	S	U.S.A. (1967)	G.A.
				7	0.11	S		
				14	0.16	S		
				21	0.05	S		
Fruits	0.12 %	EC	2	0	3.4	S	U.S.A. (1967)	G.A.
				7	0.2	S		
				14	0.22	S		
				21	0.16	S		
Blackberries								
Fruits	1.1 kg.	WP	2	0	0.59	S	U.S.A. (1961)	G.A.
				3	0.10	S		
				7	0.03	S		
				14	0.07	S		
Fruits	0.03 %	EC	1	1	0.08	S	U.S.A. (1962)	G.A.
				4	<0.05	S		
				5	<0.05	S		
Fruits	0.06 %	EC	1	1	0.23	S	U.S.A. (1962)	G.A.
				4	0.05	S		
				5	<0.05	S		
Blueberries								
Fruits	0.03 %	WP	1	0	0.19	S	U.S.A. (1961)	G.A.
				1	<0.05	S		
				2	<0.05	S		
Fruits	0.06 %	WP	1	0	0.95	S	U.S.A. (1961)	G.A.
				1	<0.05	S		
				2	<0.05	S		

Table II. (*Continued*)

Sample	Dosage (% a.i. or kg. a.i./ha.)	Formulation[a]	No. of applications	Days after last application	Residue (p.p.m.)[b]	Analytical method[c]	Country and year	Reference[d]
Boysenberries								
Fruits	0.03 %	EC	1	1	0.05	S	U.S.A. (1962)	G.A.
				4	<0.05	S		
				5	<0.05	S		
Fruits	0.06 %	EC	1	1	0.19	S		
				4	<0.05	S		
				5	<0.05	S		
Cherries								
Fruits	0.025 %	EC	1	0	1.3	GC	Switzerland (1966)	G.B.
				3	0.4	GC		
				5	<0.05	GC		
				10	0.09	GC		
				13	<0.05	GC		
Fruits	0.025 %	EC	2	0	1.5	GC	Switzerland (1966)	G.B.
				3	0.5	GC		
				5	0.3	GC		
				7	0.2	GC		
				10	0.2	GC		
				14	0.1	GC		
				21	0.04	GC		
				26	0.02	GC		
Fruits	0.025 %	EC	3	0	2.1	GC	Switzerland (1966)	G.B.
				3	0.8	GC		
				5	0.4	GC		
				7	0.4	GC		
				10	0.3	GC		
				14	0.05	GC		
				21	0.02	GC		
Filberts								
Nuts	2.2 kg.	WP	2	0	0.01	S	U.S.A. (1960)	G.A.
				3	0.01	S		
				7	0.006	S		
Shells	2.2 kg.	WP	2	0	0.078	S		
				3	0.010	S		
				7	0.000	S		
Grapes								
Fruits	1.1 kg.	WP	1	1	0.43	S	U.S.A. (1965)	G.A.
				3	0.15	S		
				7	0.06	S		
				10	0.15	S		
				14	0.07	S		
Fruits	1.1 kg.	EC	1	1	0.64	S	U.S.A. (1965)	G.A.
				3	0.23	S		
				7	0.09	S		
				10	0.14	S		
				14	0.18	S		
Grapefruit								
Peel	1.1 kg.	WP	1	7	0.11	S	U.S.A. (1961)	G.A.
				14	0.11	S		
				21	0.24	S		
Pulp	1.1 kg.	WP	1	7	<0.05	S		
				14	<0.05	S		
				21	<0.05	S		
Peel	2.2 kg.	WP	1	7	0.21	S	U.S.A. (1961)	G.A.
				14	0.11	S		
				21	0.18	S		
Pulp	2.2 kg.	WP	1	7	<0.05	S		
				14	<0.05	S		
				21	<0.05	S		

Table II. (*Continued*)

Sample	Dosage (% a.i. or kg. a.i./ha.)	Formulation[a]	No. of applications	Days after last application	Residue (p.p.m.)[b]	Analytical method[c]	Country and year	Reference[d]
Lemons								
Peel	0.06 %	WP	1	1	11.6**	Sp	U.S.A. (1956)	GUNTHER *et al.* (1958)
				4	4.3	Sp		
				7	2.6	Sp		
				14	2.6	Sp		
				21	1.1	Sp		
				28	0.8	Sp		
Pulp	0.06 %	WP	1	Various	ND	Sp	U.S.A. (1956)	GUNTHER *et al.* (1958)
Limes								
Peel	0.3 kg.	WP	1	1	0.39	S	U.S.A. (1962)	G.A.
				8	0.31	S		
				14	<0.05	S		
Pulp	0.3 kg.	WP	1	1	0.27	S	U.S.A. (1962)	G.A.
				8	<0.05	S		
				14	<0.05	S		
Peel	0.6 kg.	WP	1	1	0.61	S	U.S.A. (1962)	G.A.
				8	0.29	S		
				14	0.34	S		
Pulp	0.6 kg.	WP	1	1	<0.05	S	U.S.A. (1962)	G.A.
				8	<0.05	S		
				14	<0.05	S		
Nectarines								
Fruits	0.06 %	EC	2	0	2.8	S	U.S.A. (1967)	G.A.
				7	0.64	S		
				10	0.32	S		
				14	0.25	S		
Oranges								
Peel	0.06 %	WP	1	0	3.5**	Sp	U.S.A. (1956)	GUNTHER *et al.* (1958)
				1	1.9	Sp		
				4	0.5	Sp		
				7	0.5	Sp		
				14	0.4	Sp		
				21	0.3	Sp		
				28	0.3	Sp		
Pulp	0.06 %	WP	1	Various	ND	Sp		
Peel	0.06 %	EC	1	0	4.0**	Sp	U.S.A. (1956)	GUNTHER *et al.* (1958)
				1	1.1	Sp		
				4	0.9	Sp		
				7	0.9	Sp		
				14	0.5	Sp		
				21	0.2	Sp		
				28	0.3	Sp		
Pulp	0.06 %	EC	1	Various	ND	Sp		
Peaches								
Fruits	0.06 %	EC	4	0	3.9	S	U.S.A. (1967)	G.A.
				7	0.71	S		
				12	0.38	S		
				15	0.23	S		
Fruits	0.04 %	EC	1	14	0.02	GC	Italy (1969)	G.B.
Pears								
Fruits	0.06 %	WP	1	1	0.513	S	U.S.A. (1955)	G.A.
				9	0.513	S		
				15	0.002	S		
Fruits	0.04 %	EC	1	14	0.02	GC	Italy (1969)	G.B.
Plums								
Fruits	0.06 %	EC	2	0	1.17	S	U.S.A. (1967)	G.A.
				6	0.06	S		
				11	<0.05	S		
				15	<0.05	S		

Table II. (*Continued*)

Sample	Dosage (% a.i. or kg. a.i./ha.)	Formulation[a]	No. of applications	Days after last application	Residue (p.p.m.)[b]	Analytical method[c]	Country and year	Reference[d]
Plums (*Cont.*)								
Fruits	0.12 %	EC	2	0	3.39	S	U.S.A. (1967)	G.A.
				6	0.19	S		
				11	0.06	S		
				15	<0.05	S		
Prunes								
Fruits	0.6 kg.	EC	3	0	1.35	S	U.S.A. (1967)	G.A.
				6	0.13	S		
				9	<0.05	S		
				14	<0.05	S		
Fruits	1.1 kg.	EC	3	0	2.9	S	U.S.A. (1967)	G.A.
				6	0.31	S		
				9	0.26	S		
				14	0.08	S		
				21	0.05	S		
Strawberries								
Fruits	0.06 %	EC	2	20	0.20	GC	Switzerland (1966)	G.B.
				27	<0.05	GC		
				36	<0.05	GC		
				55	<0.05	GC		
Walnuts								
Nuts	3.4 kg.	WP	2	28	<0.05	S	U.S.A. (1966)	G.A.
Shells	3.4 kg.	WP	2	28	<0.05	S		

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[a] WP = wettable powder, EC = emulsive concentrate, and G = granular formulation.

[b] Single star-average of two analyses, double star = average of three analyses, and ND = none detected.

[c] GC = gas chromatographic method, S = sulfide method, Sp = spectrophotometric method, Ph = phosphorus method, and ChI = cholinesterase inhibition method (see preceding review, this volume).

[d] G.A. = Geigy Chemical Corporation, Ardsley, New York, U.S.A.; G.B. = J. R. Geigy S. A., Basle, Switzerland; and M.-B. = personal communication from Professor H. MAIER-BODE, Institute of Pharmacology, University of Bonn, G.F.R.

[MOSEBACH and STEINER 1960, TIITTANEN and VARIS 1964 (cited by FINLAYSON *et al.* 1968), SUETT 1971]. In the studies with carrots residue levels varied widely depending on formulation, method of application, and time of treatment. They varied particularly with granular formulations applied to the soil, giving probably a rather uneven distribution. However, all trials performed in Switzerland indicated that 60 days after the last treatment as a soil application residues in carrots were below 0.5 p.p.m. in all cases. According to RYGG and SØMME (1967), diazinon should not be recommended for early harvest carrots. The uptake of diazinon by carrots was found to depend on the soil type; in no case was it constant during the time between treatment and harvest (FINLAYSON *et al.* 1968).

Other root vegetables (radishes, rutabagas, onions) also treated mainly with granular formulations showed no unacceptable residues.

Residues in beans, peas, cucumbers, and tomatoes were very low (0.1 p.p.m. after 14 days).

Table III. *Residues of diazinon in vegetables.*

Sample	Dosage (% a.i. or kg. a.i./ha.)	Formulation[a]	No. of applications	Days after last application	Residue (p.p.m.)[b]	Analytical method[c]	Country and year	Reference[d]
Beans (pole)								
Pods	0.02 %	EC	1	0	0.2	S	Germany G.F.R. (1961)	STOBWASSER (1963)
				3	0.05	S		
Pods	0.06 %	EC	1	0	0.70	S	Germany G.F.R. (1961)	STOBWASSER (1963)
				3	0.20	S		
				7	<0.03	S		
Beans (snap)								
Pods	0.4 kg.	EC	1	7	0.14*	S	U.S.A. (1965)	G.A.
				14	0.05	S		
Pods	0.8 kg.	EC	1	7	0.17	S		
				14	0.05	S		
Pods	1.7 kg.	EC	1	7	0.37	S		
				14	0.07	S		
Broccoli								
Heads	0.6 kg.	EC	3	0	0.74*	S	U.S.A. (1966)	G.A.
				3	0.07	S		
				5	0.06	S		
				7	0.06	S		
				10	<0.05	S		
Marketable sprouts	0.6 kg.	EC	2	0	1.2*	S	U.S.A. (1967)	G.A.
				3	0.2	S		
				7	0.08	S		
				10	0.07	S		
				14	<0.05	S		
Brussels sprouts								
Heads	0.6 kg.	EC	1	0	1.9*	GC	U.S.A. (1965)	G.A.
				3	0.62	GC		
				7	0.46	GC		
				14	0.34	GC		
Heads	0.6 kg.	EC	2	0	0.11*	GC	U.S.A. (1966)	G.A.
				3	0.14	GC		
				7	<0.10	GC		
Heads	1.1 kg.	EC	2	0	0.29*	GC		
				3	0.12	GC		
				7	<0.10	GC		
Cabbage								
Head	0.025 % (drench)	EC	1	0	12.5	GC	Switzerland (1966)	G.B.
				2	0.8	GC		
				7	1.1	GC		
				14	0.2	GC		
				21	<0.05	GC		
				28	<0.05	GC		
Head	0.025 % (drench)	EC	1	0	27.0	GC	Switzerland (1966)	G.B.
				2	11.0	GC		
				7	1.5	GC		
				14	0.4	GC		
				21	0.1	GC		
				28	0.08	GC		
				35	<0.05	GC		
Entire head	0.6 kg.	EC	2	0	1.08*	S	U.S.A. (1967)	G.A.
				3	0.65	S		
				7	0.16	S		
				10	0.14	S		
				14	0.23	S		
Trimmed head	0.6 kg.	EC	2	0	0.10*	S	U.S.A. (1967)	G.A.
				3	0.07	S		
				7	0.10	S		
				10	<0.05	S		
				14	0.12	S		

Table III. (*Continued*)

Sample	Dosage (% a.i. or kg. a.i./ha.)	Formulation[a]	No. of applications	Days after last application	Residue (p.p.m.)[b]	Analytical method[c]	Country and year	Reference[d]
Carrots								
Roots	1.8 kg. (drilled)	G	1	112	0.34*	S	Germany G.F.R. (1963)	G.B.
Roots	0.025 % (drench)	EC	1	112	3.2*	S	Germany G.F.R. (1963)	G.B.
Roots	0.025 % (drench)	EC	1	19	1.7	GC	Switzerland (1966)	G.B.
				42	0.2	GC		
				56	0.08	GC		
				70	0.03	GC		
Roots	0.025 % (drench)	EC	2	2	2.2	GC	Switzerland (1966)	G.B.
				16	1.8	GC		
				30	0.7	GC		
Roots	4 kg. (in band)	G	2	29	1.3	GC	Switzerland (1966)	G.B.
				45	1.2	GC		
				59	0.5	GC		
				73	0.2	GC		
Tops	0.025 % (drench)	EC	1	19	1.0	GC	Switzerland (1966)	G.B.
				28	1.0	GC		
				35	0.5	GC		
Roots	2 kg. (drilled)	G	1	97	0.14	GC	Sweden (1966)	G.B.
	2.5 kg. (drilled)	G	1	97	0.40	GC		
Total root	3.65 kg	EC	1	61	0.34	ChI	Belgium (1969)	STATHOPOULOS *et al.* (1971)
				81	0.14	ChI		
				91	0.07	ChI		
				130	<0.05	ChI		
Peel	3.65 kg	EC	1	91	0.35	ChI		
				130	0.05	ChI		
				188	<0.05	ChI		
Cauliflower								
Heads	0.6 kg.	EC	3	0	0.94*	S	U.S.A. (1966)	G.A.
				3	0.42	S		
				5	0.32	S		
				7	0.19	S		
				10	0.29	S		
Heads	0.04 %	WP	1	21	0.016	GC	Germany G.F.R. (1968)	M.—B.
Celery								
Above ground parts	0.6 kg.	EC	1	0	4.8	S	U.S.A. (1965/66)	G.A.
				3	1.1	S		
				7	1.5	S		
				10	0.7	S		
				28	0.79	S		
				35	0.13	S		
Tuberous base	6.3 kg.	G	1	97	0.41	GC	Switzerland (1968)	G.B.
Tuberous base	9.45 kg.	G	1	97	0.85	GC		
Collards								
Green leaves	0.6 kg.	EC	2	0	3.9*	S	U.S.A. (1967)	G.A.
				3	1.02	S		
				7	0.38	S		
				10	0.27	S		
				14	0.21	S		
Cucumber								
Fruits	0.03 %	WP	1	1	0.10	S	U.S.A. (1967)	G.A.
				2	0.07	S		
				4	0.05	S		
				8	0.17	S		

Table III. (*Continued*)

Sample	Dosage (% a.i. or kg. a.i./ha.)	Formulation[a]	No. of applications	Days after last application	Residue (p.p.m.)[b]	Analytical method[c]	Country and year	Reference[d]
Cucumber (*Cont.*)								
Fruits	0.06 %	WP	1	1	0.16	S	U.S.A. (1967)	G.A.
				2	0.17	S		
				4	0.05	S		
				8	0.13	S		
Fruits (greenhouse)	0.3 kg.	EC	1	0	0.17	S	U.S.A. (1967)	G.A.
				1	0.18	S		
				4	0.39	S		
				8	0.15	S		
Fruits (greenhouse)	0.6 kg.	EC	1	0	1.0	S		
				1	1.2	S		
				4	0.82	S		
				8	0.26	S		
Fruits (under glass)	0.02 %	EC	1	0	0.05	S	Germany G.F.R. (1961)	STOBWASSER (1963)
				3	0.03	S		
Fruits (under glass)	0.03 %	EC	1	0	0.35	S	Germany G.F.R. (1961)	STOBWASSER (1963)
				3	0.15	S		
				7	0.10	S		
Endive								
Foliage	0.3 kg.	EC	2	0	17.5*	S	U.S.A. (1966)	G.A.
				3	10.0	S		
				7	3.55	S		
				10	2.40	S		
				14	2.10	S		
Kale								
Foliage	0.3 kg.	EC	2	0	6.3	S	U.S.A. (1966)	G.A.
				3	2.0	S		
				7	0.58	S		
				10	0.31	S		
				14	0.17	S		
Leeks	0.025 %	EC	3	55	<0.02	GC	Switzerland (1966)	G.B.
Lettuce								
Foliage	0.6 kg.	EC	2	0	4.0*	S	U.S.A. (1967)	G.A.
				3	2.25	S		
				7	0.59	S		
				10	0.16	S		
				14	0.09	S		
Heads (field applied)	0.02 %	EC	1	0	1.5	S	Germany G.F.R. (1961)	STOBWASSER (1961)
				7	0.05	S		
				14	<0.03	S		
Heads (under glass)	0.02 %	EC	1	0	4.8	S		
				8	0.21	S		
				14	0.05	S		
Heads	0.5 kg.		1	0	8.1	Ph	Canada (1964)	COFFIN & KINLEY (1964)
				2	4.0	Ph		
				7	0.3	Ph		
				10	<0.1	Ph		
Onions								
Bulbs and tops	11.2 kg.	G	1	12	<0.2	S	U.S.A. (1965)	G.A.
Bulbs and tops	16.8 kg.	G	1	12	<0.2	S		
Green bulbs	11.2 kg.	EC	1	133	<0.1	GC	U.S.A. (1967)	G.A.
Green bulbs	16.8 kg.	G	1	133	<0.1	S		
Bulbs	2 kg.	G	1	125	<0.05	GC	Switzerland (1967)	G.B.
Bulbs	4 kg.	G	1	137	<0.05	GC		
Bulbs	10 kg. (drench)	EC	1	137	<0.05	GC		
Parsley	0.6 kg.	EC	2	0	6.6*	GC	U.S.A. (1966/67)	G.A.
				3	4.5	GC		
				7	4.45	GC		

Table III. (*Continued*)

Sample	Dosage (% a.i. or kg. a.i./ha.)	Formulation[a]	No. of applications	Days after last application	Residue (p.p.m.)[b]	Analytical method[c]	Country and year	Reference[d]
Parsley (Cont.)								
				10	2.70	GC		
				14	2.10	GC		
Peas								
Vines and pods	0.06 %	EC	1	0	1.45	S	U.S.A. (1958)	G.A.
				2	0.18	S		
				4	0.15	S		
				8	0.05	S		
				15	0.13	S		
Radishes								
Roots	4.5 kg.	EC	1	82	<0.05	S	U.S.A. (1963)	G.A.
Roots	4.5 kg.	G	1	82	<0.05	S		
Roots	9.0 kg.	EC	1	82	<0.05	S		
Roots	9.0 kg.	G	1	82	<0.05	S		
Roots	3 kg.	G	1	34	0.06	GC	Switzerland (1967)	G.B.
				36	<0.02	GC		
Roots	10 kg.	EC	1	34	<0.02	GC		
Rutabagas								
Roots	2 kg. +3 × 0.9 kg.	G EC	4	30	0.09	S	U.S.A. (1967)	G.A.
	2 kg. +3 × 0.9 kg.	G EC	4	30	0.09	S		
Spinach								
Leaves	0.025 %	EC	1	2	1.1	GC	Switzerland (1967)	G.B.
			1	7	0.02	GC		
			1	12	<0.01	GC		
Canned, processed Spinach	1.5 kg.		1	5	<0.1*	S	U.S.A. (1967)	G.A.
Water	1.5 kg.		1	5	<0.1	S		
Tomatoes								
Fruits	0.03 %	EC	1	1	2.5	S	U.S.A. (1966/67)	G.A.
				2	1.9	S		
				4	0.77	S		
				8	0.40	S		
Fruits	0.06 %	WP	1	1	2.6	S		
				2	1.4	S		
				4	0.54	S		
				8	0.43	S		
Greenhouse fruits	0.03 %	EC	1	1	0.54	S	U.S.A. (1966)	G.A.
				5	0.15	S		
				7	0.19	S		
				14	0.15	S		
Greenhouse fruits	0.06 %	EC	1	1	0.84	S		
				5	0.83	S		
				7	0.15	S		
				14	0.17	S		
Turnips								
Tops	0.6 kg.	EC	2	0	5.7*	S	U.S.A. (1966)	G.A.
				3	3.7	S		
				7	2.4	S		
				10	1.35	S		
				14	0.59	S		

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[a] WP = wettable powder, EC = emulsive concentrate, and G = granular formulation.

[b] Single star = average of two analyses and double star = average of three analyses.

[c] GC = gas chromatographic method, S = sulfide method, Sp = spectrophotometric method, Ph = phosphorus method, and ChI = cholinesterase inhibition method (see preceding review, this volume).

[d] G.A. = Geigy Chemical Corporation, Ardsley, New York, U.S.A.; G.B. = J. R. Geigy S. A., Basle, Switzerland; and M.-B. = personal communication from Professor H. MAIER-BODE, Institute of Pharmacology, University of Bonn, G.F.R.

Leafy vegetables had relatively high initial residues which, however, dissipated rapidly. Thus, the half-life value on lettuce, spinach, collards, broccoli, kale, celery, etc. was only two days. Like other insecticides, diazinon was also more stable on crops under glass: STOBWASSER and KIRCHHOFF (1966) found four times higher residues on greenhouse lettuce as compared to field-treated lettuce at equal application rates.

1. Distribution. — Tracer studies showed that diazinon translocated rapidly and unchanged in bean plants; thus, 18 hours after application to the above-ground parts the compound was detected in the root exudates (GUNNER *et al.* 1966). Translocation also occurred in the opposite direction; thus, pea plants grown in a soil with 25 p.p.m. of labelled diazinon had, after 15 days, radioactivity in their roots and in their green parts corresponding to 9.45 and 0.27 p.p.m., respectively (LICHTENSTEIN *et al.* 1967). Acropetal transport in peas was also demonstrated by LAANIO *et al.* (1972). Additionally, KANSOUH and HOPKINS (1968) observed that diazinon passed freely into and out of the roots of beans in hydroponic solution.

2. Metabolites. — In peas unchanged diazinon represented the main part of the radioactivity uptake by roots in hydroponic solution but efficient degradation took place in the shoots; "pyrimidinol" and "hydroxy-pyrimidinol" were the main products found (LAANIO *et al.* 1972). Two days after placement of beans in a hydroponic solution containing labelled diazinon the unchanged compound was found only in the primary leaves. After longer time intervals, the terminal leaves and foliage contained exclusively "pyrimidinol" and other metabolites extractable with chloroform. Cleavage of the pyrimidine ring was slow, with only about 0.1 percent of the absorbed radioactivity being evolved as $^{14}CO_2$ during the first six days. C^{14}-diazoxon was not detected, indicating that the oxidation was very slow or that the diazoxon was hydrolyzed as rapidly as it was formed (KANSOUH and HOPKINS 1968, EBERLE and NOVAK 1969).

On field-sprayed lettuce neither diazoxon nor other metabolites were found within 14 days after treatment (COFFIN and MCKINLEY 1964).

RALLS *et al.* (1966) found a rapid decrease in diazinon residues in an extensive field study carried out with ^{35}S-labelled diazinon in spinach, tomatoes, and snap beans. Diazoxon was the only detectable metabolite with cholinesterase-inhibiting properties in spinach; it ranged from 0.005 to 0.001 p.p.m. five days after spraying. Snap beans also seemed to contain a faint amount of diazoxon seven days after treatment. "Pyrimidinol" was identified in tomatoes five days after spraying with pyrimidine ring-labelled ^{14}C-diazinon.

"Hydroxy-diazinon" was detected in very small amounts in field-sprayed kale (PARDUE *et al.* 1970) and, in a greenhouse experiment, in beans (*CIBA-GEIGY* 1972 b) (see Table IV).

Table IV. *Residues of diazinon and its metabolites in kale and beans.*

Crop and a.i. applied	Interval (days)	Residue (p.p.m.)		
		Diazinon	Diazoxon	"Hydroxy-diazinon"
Kale	2	8.8	0.004	0.18
(2.2 kg./ha.)	7	2.9	0.007	0.05
	11	2.0	0.002	0.03
	15	1.6	0.002	0.03
Beans	1	33.5	0.011	0.17
(0.05% to run-off)	3	15.7	0.012	0.12
	7	3.9	<0.018	0.08
	14	0.71	<0.008	0.06
	21	0.16	<0.008	0.04
Limit of detection	—	0.05	0.008	0.025

c) *Field crops* (Table V)

No residues of diazinon could be detected in the grains of corn, oat, sorghum, rice, and wheat or in the tubers of potatoes after application to the established crops at various stages by emulsions or granules.

The seeds of cotton, safflowers or sunflowers contained only little diazinon, in contrast to the oil obtained from these seeds. Thus only 0.07 p.p.m. of diazinon was detected in harvested sunflower seeds, even when the last application was made 14 days before sampling (Randolph *et al.* 1969). The amount of diazinon in refined oil prepared from cottonseeds containing 0.05 p.p.m. 21 days after treatment was found to be 0.15 p.p.m.

The tolerances for diazinon in corn, sorghum, peanuts, soybeans, sugarcane, and pineapple are established according to the use of these crops either as food or feed. The residue levels obtained show that the established tolerances are by no means exceeded.

1. Distribution. — According to Onsager and Rusk (1967), sugarbeet seedlings absorb soil-applied diazinon in insecticidal quantities: 0.32 p.p.m. were detected in the foliage after a pre-plant granular application. In beans the absorbed amount was too low to exert any insecticidal activity (Gunner *et al.* 1966). In a laboratory study, rice plants transpired diazinon applied to the paddy water in insecticidal quantities (Laanio *et al.* 1972).

2. Metabolites. — In the study by Laanio *et al.* (1972) unchanged diazinon was the main part of the radioactivity taken up during the first days. Later the radioactivity in the shoots consisted of small

Table V. *Residues of diazinon in field crops.*

Sample	Dosage (% a.i. or kg. a.i./ha.)	Formulation[a]	No. of applications	Days after last application	Residue (p.p.m.)[b]	Analytical method[c]	Country and year	Reference[d]
Corn (maize)								
Kernels	2 kg.	G	1	0	2.5	GC	Switzerland (1966)	G.B.
				2	0.16	GC		
				7	0.05	GC		
Forage	2 kg.	G	1	0	15.40	GC		
				2	5.40	GC		
				7	0.22	GC		
				14	1.1	GC		
				21	0.3	GC		
Silage stage	1.1 kg.	G	1	58	0.07*	S	U.S.A. (1961)	G.A.
Silage stage	2.2 kg.	G	1	58	0.09	S		
Leaves and stalks	1.1 kg.	EC	1	1	1.79*	S	U.S.A. (1966)	G.A.
				3	0.31	S		
				8	<0.05	S		
				15	<0.05	S		
				83	<0.05	S		
Entire plant	1.1 kg.	G	1	1	1.67	GC	U.S.A. (1969)	HARDING *et al.* (1969)
				3	0.77	GC		
				7	0.0	GC		
Leaves	1.1 kg.	G	1	1	0.43	GC		
				3	1.50	GC		
				7	0.0	GC		
Cotton								
Hulls	0.5 kg.	EC	2	1	<0.05	S	U.S.A. (1965)	G.A.
				7	<0.05	S		
Seed	0.5 kg.	EC	2	1	0.06	S		
				7	<0.05	S		
Refined oil	0.5 kg.	EC	2	1	0.10	S		
				7	<0.05	S		
Hulls	1.2 kg.	EC	2	1	0.3	S		
				7	0.19	S		
				21	0.06	S		
Seed	1.2 kg.	EC	2	1	0.2	S		
				7	0.08	S		
				21	0.05	S		
Refined oil	1.2 kg.	EC	2	1	0.32	S		
				7	0.16	S		
				21	0.15	S		
Oats								
Tops	1.2 kg.	EC	1	40	0.07	S	U.S.A. (1961)	G.A.
Bottoms	1.2 kg.	EC	1	40	0.07	S		G.A.
Peanuts								
Meat	1.1 kg.	G	1	64	<0.05*	S	U.S.A. (1962)	G.A.
Shells	1.1 kg.	G	1	64	0.5	S		
Hay	1.1 kg.	G	1	64	0.10	S		
Meat	2.2 kg.	G	1	64	<0.05	S		
Shells	2.2 kg.	G	1	64	0.41	S		
Meat	5.6 kg.	G	1	20	0.19*	S	U.S.A. (1965)	G.A.
Shells	5.6 kg.	G	1	20	1.5	S		
Meat	11.0 kg.	G	1	20	0.28	S		
Meat	11.0 kg.	G	1	42	0.22	S		
Shells	11.0 kg.	G	1	20	4.4	S		
	11.0 kg.	G	1	42	4.5	S		
Pineapples								
Foliage	4.5 kg.	EC	1	0	77.0*	S	Hawaii (1965)	G.A.
				3	8.0	S		
				7	6.8	S		

Table V. (*Continued*)

Sample	Dosage (% a.i. or kg. a.i./ha.)	Formulation[a]	No. of applications	Days after last application	Residue (p.p.m.)[b]	Analytical method[c]	Country and year	Reference[d]
Pineapples (Cont.)								
Rind	4.5 kg.	EC	1	0	1.38	S		
				3	0.23	S		
				7	0.21	S		
Meat	4.5 kg.	EC	1	0	0.06	S		
				3	<0.05	S		
				7	<0.05	S		
Potatoes								
Skin	4.5 kg.	G	1	42	0.16*	S	U.S.A. (1965)	G.A.
Meat	4.5 kg.	G	1	42	0.05	S		
Skin	9.0 kg.	G	1	42	0.08	S		
Meat	9.0 kg.	G	1	42	0.05	S		
Skin	5.2 kg.	G	1	50	<0.05	S	U.S.A. (1965)	G.A.
Meat	5.2 kg.	G	1	50	<0.05	S		
Skin	10.4 kg.	G	1	50	<0.05	S		
Meat	10.4 kg.	G	1	50	<0.05	S		
Sweet potatoes								
Skin	4.5 kg.	G	1	167	<0.05	S	U.S.A. (1964)	G.A.
Meat	4.5 kg.	G	1	167	<0.1	S		
Rice								
Polished grains	3.0 kg.	G	1	105	<0.05	GC	Phillipines (1966)	G.B.
Straw	3.0 kg.	G	1	105	<0.05	GC		
Polished grains	2.0 kg.	G	1	105	<0.05	GC		
Straw	2.0 kg.	G	1	105	<0.05	GC		
Safflower								
Seeds	1.65 kg.	EC	2	32	<0.05*	GC	U.S.A. (1967)	G.A.

amounts of the polar metabolites "pyrimidinol" and "hydroxy-pyrimidinol". "Hydroxy-diazinon" was only a minor metabolite, not exceeding two percent of the total radioactivity absorbed by the rice plants. The same pattern of metabolites was found after stem injection; diazoxon did not exceed 0.01 percent of the extractable radioactivity. No diazoxon was found in field corn treated with three different formulations of diazinon (HARDING *et al.* 1969).

d) Forage crops (Table VI)

Immediately after treatment of forage crops (alfalfa, clover, grass, cowpeas, lespedeza) residues of diazinon are rather high (>50 p.p.m.), but they decline rapidly. The estimated half-life of diazinon in alfalfa and grasses was less than two days. Residue determinations in cattle (FAO/WHO 1971) indicate that no residues occur in meat and milk if the animals are fed forage treated according to label instructions.

Table V. (*Continued*)

Sample	Dosage (% a.i. or kg. a.i./ha.)	Formulation[a]	No. of applications	Days after last application	Residue (p.p.m.)[b]	Analytical method[c]	Country and year	Reference[d]
Sorghum								
Grains	0.8 kg.	EC	1	0	0.46	S	U.S.A. (1963)	G.A.
				3	0.06	S		
				6	<0.05	S		
				14	<0.05	S		
Foliage	0.8 kg.	EC	1	0	3.5	S		
				3	0.13	S		
				6	0.22	S		
				14	<0.05	S		
Stalks	0.6 kg.	EC	1	0	2.0*	S	U.S.A. (1967)	G.A.
				3	2.1	S		
				7	2.9	S		
				23	0.10	S		
				32	0.16	S		
Grains	0.6 kg.	EC	1	32	<0.05	S		
Stalks	0.6 kg. (airial appl.)	EC	1	0	1.6	S		
				3	1.3	S		
				7	5.1	S		
				23	0.05	S		
				32	0.16	S		
Grains	0.6 kg.	EC	1	32	<0.05	S		
Soybeans								
Whole plant	3.4 kg.	G	1	30	0.08	S	U.S.A. (1966)	G.A.
Whole plant	6.7 kg.	G	1	30	0.09	S		
Foliage	1.1 kg.	EC	1	1	11.0	S	U.S.A. (1966)	G.A.
				8	3.1	S		
				15	1.8	S		
				22	0.8	S		
Foliage	2.2 kg.	EC	1	1	0.35	S		
				8	0.11	S		
				15	0.09	S		
				22	0.84	S		
Sugarcane								
Stems	0.6 kg.	EC	1	0	0.1	S	U.S.A. (1962)	G.A.
				3	<0.05	S		
				7	<0.05	S		
				14	<0.05	S		
Leaves	0.6 kg.	EC	1	0	4.05	S		
				3	0.12	S		
				7	0.06	S		
				14	<0.05	S		
Sugarcane, processed								
Molasses	3.0 kg.	G	1	28	0.06	S	U.S.A. (1964)	G.A.
Syrup	3.0 kg.	G	1	28	0.08	S		
Raw sugar	3.0 kg.	G	1	28	<0.05	S		
Clarified juice	3.0 kg.	G	1	28	<0.05	S		
Mud from clarifiers	3.0 kg.	G	1	28	0.16	S		
Bagasse	3.0 kg.	G	1	28	0.72	S		
Sunflower								
Seeds	1.1 kg.	EC	1	88	0.05	GC	U.S.A. (1964)	G.A.
Seeds	1.1 kg.	EC	1	88	0.13	S		
Oil	1.1 kg.	EC	1	88	0.20	GC		
Oil	1.1 kg.	EC	1	88	0.19	S		
Oil	1.1 kg.	EC	2	81	0.46	GC		
Oil	1.1 kg.	EC	2	81	0.51	S		

Table V. (*Continued*)

Sample	Dosage (% a.i. or kg. a.i./ha.)	Formulation[a]	No. of applications	Days after last application	Residue (p.p.m.)[b]	Analytical method[c]	Country and year	Reference[d]
Sunflower (Cont.)								
Seeds	1.65 kg.	EC	3	24	0.43*	GC	U.S.A. (1967)	G.A.
				38	1.03	GC		
				52	1.30	GC		
				66	0.88	GC		
Seeds	1.1 kg.	EC	2	41	0.03*	GC	U.S.A. (1967)	Randolph *et al.* (1969)
			4	14	0.07	GC		
Wheat								
Entire plant	0.6 kg.	EC	1	0	4.9*	S	U.S.A. (1965)	G.A.
				6	0.5	S		
				13	0.1	S		
Grain	0.6 kg.	EC	1	20	<0.05	S		
Entire plant	1.1 kg.	EC	1	0	7.3	S		
				6	0.6	S		
				13	0.2	S		
Grain	1.1 kg.	EC	1	20	<0.05	S		

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[a] WP = wettable powder, EC = emulsive concentrate, and G = granular formulation.

[b] Single star = average of two analyses and double star = average of three analyses.

[c] GC = gas chromatographic method, S = sulfide method, Sp = spectrophotometric method, Ph = phosphorus method, and ChI = cholinesterase inhibition method (see preceding review, this volume).

[d] G.A. = Geigy Chemical Corporation, Ardsley, New York, U.S.A.; G.B. = J. R. Geigy S. A., Basle, Switzerland; and M.-B. = personal communication from Professor H. Maier-Bode, Institute of Pharmacology, University of Bonn, G.F.R.

1. Distribution. — During 24 hours diazinon was absorbed by alfalfa grown in a greenhouse soil treated with the insecticide. Since the amounts of residues remained constant during nine to 15 days after treatment, they might depend to a great extent on the concentration of diazinon in the soil. Therefore, the stabilization or decrease of diazinon levels in the plant was due to the apparent equilibrium between diazinon residues in the plants and the soil and the loss of diazinon by transpiration.

2. Metabolites. — No metabolites, not even diazoxon, were detected in the samples analyzed (Nelson and Hamilton 1970); however, the data presented do not preclude the possibility that metabolism could have occurred.

e) *Various plant products* (Table VII)

Residues in green coffee beans and tea leaves were low. In fermented coffee beans and brewed tea diazinon could not be detected.

The dissipation of diazinon in tobacco was not accelerated by curing; rather, the concentration increased during this process.

1. Distribution. — Due to the solubility characteristic of diazinon, residues tend to persist in oil fruits and to concentrate in the oil

Table VI. *Residues of diazinon on forage crops.*

Sample	Dosage (% a.i. or kg. a.i./ha.)	Formulation[a]	No. of applications	Days after last application	Residue (p.p.m.)[d]	Analytical method[b]	Country and year	Reference[d]
Alfalfa								
Foliage	1.1 kg.	EC	2	1	15.57	S	U.S.A. (1961)	G.A.
				3	6.57	S		
				5	2.80	S		
				15	0.17	S		
Fresh foliage	0.6 kg.	EC	1	0	11.0*	S	U.S.A. (1963)	G.A.
				3	0.69	S		
				7	0.21	S		
				14	0.05	S		
Fresh foliage	0.8 kg.	EC	1	0	25.5	S		
				3	1.9	S		
				7	0.26	S		
				14	0.15	S		
Air-dried foliage	0.6 kg.	EC	1	0	17.5	S		
				3	0.38	S		
				7	0.10	S		
				14	0.11	S		
Air-dired foliage	0.8 kg.	EC	1	0	54.5	S		
				3	3.4	S		
				7	0.36	S		
				14	0.11	S		
Beets								
Foliage	8 + 10 kg.	G	2	90	0.5	S	Switzerland (1962)	G.B.
Foliage	4 + 10 kg.	G	2	90	0.1	S		
Foliage	8 + 10 kg.	G	2	90	2.8	S		
Roots	8 + 10 kg.	G	2	90	0.2	S		
Roots	4 + 10 kg.	G	2	90	<0.05	S		
Roots	8 + 10 kg.	G	2	90	0.1	S		
Clover								
Red	0.6 kg.	EC	1	0	9.43	S	U.S.A. (1962)	G.A.
				1	4.53	S		
				7	0.78	S		
				14	0.26	S		
Red	1.1 kg.	EC	1	0	34.83	S		
				1	8.67	S		
				7	2.13	S		
				14	1.34	S		
Ladino	1.1 kg.	EC	2	1	28.4	S	U.S.A. (1961)	G.A.
				3	6.9	S		
				5	5.24	S		
				15	0.24	S		
Cowpeas								
Whole plant	3.4 kg.	G	1	30	0.07	S	U.S.A. (1966)	G.A.
Whole plant	6.7 kg.	G	1	30	0.06	S		
Forage	3.4 kg.	G	1					
	+1.1 kg.	EC	1	0	17.0	S	U.S.A. (1966)	G.A.
				7	11.0	S		
				14	6.1	S		
				21	4.2	S		
Forage	3.4 kg.	G	1					
	+2.2 kg.	EC	1	0	34	S	U.S.A. (1966)	G.A.
				7	13	S		
				14	11	S		
				21	7.1	S		
Grass								
(under apple trees)	0.05 %	EC	1	0	12.0	S	Germany G.F.R. (1963)	MAIER-BODE (1963)
				3	0.4	S		
				10	<0.1	S		
				17	<0.1	S		

Table VI. (*Continued*)

Sample	Dosage (% a.i. or kg. a.i./ha.)	Formulation[a]	No. of applications	Days after last application	Residue (p.p.m.)[d]	Analytical method[b]	Country and year	Reference[d]
Grass Cont.								
Hay				23	<0.1	S		
Tops	1.1 kg.	WP	1	0	20.0	S	Puerto Rico (1965)	G.A.
				3	6.3	S		
				7	0.20	S		
Tops	2.2 kg.	WP	1	0	23.0	S		
				3	2.5	S		
				7	0.08	S		
Bermuda grass tops	1.1 kg.	EC	1	0	10.5	S	U.S.A. (1965)	G.A.
				3	0.76	S		
Bermuda grass tops	2.2 kg.	EC	1	0	22.5	S		
				3	4.3	S		
Lespedeza, above-ground part	3.4 kg.	G	1	45	0.28	S	U.S.A. (1966)	G.A.
Lespedeza, above-ground part	6.7 kg.	G	1	45	0.27	S		
Lespedeza, above-ground part	3.4 kg. (pre-plant)	G	1	30	<0.05	S	U.S.A. (1967)	G.A.
Lespedeza, above-ground part	6.7 kg. (pre-plant)	G	1	30	0.09	S		
Green forage	3.4 kg.	G	1	90	<0.05*	S		
Dried forage	3.4 kg.	G	1	90	0.08	S		
Green forage	6.7 kg.	G	1	90	<0.05	S		
Dried forage	6.7 kg.	G	1	90	0.09	S		

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[a] WP = wettable powder, EC = emulsive concentrate, and G = granular formulation.

[b] Single star = average of two analyses and double star = average of three analyses.

[c] GC = gas chromatographic method, S = sulfide method, Sp = spectrophotometric method, Ph = phosphorus method, and ChI = cholinesterase inhibition method (see preceding review, this volume).

[d] G.A. = Geigy Chemical Corporation, Ardsley, New York, U.S.A.; G.B. = J. R. Geigy S. A., Basle, Switzerland; and M.-B. = personal communication from Professor H. Maier-Bode, Institute of Pharmacology, University of Bonn, G.F.R.

fraction. However, if recommendations are observed, the residue levels in olives were below the established tolerances in Italy of two p.p.m. and in the United States of one p.p.m., seven to eight weeks after treatment.

2. Metabolites. — The only metabolite detected in olive oil was diazoxon at 0.007 p.p.m. Monothionotetraethylpyrophosphate (S-TEPP) and TEPP could not be detected in olives at any time between application and harvest (Eberle and Novak 1969).

III. Diazinon in processed and stored foodstuffs

Processing and canning operations substantially reduce the levels of diazinon residues still present on and in harvested crops.

Table VII. *Residues of diazinon on various plant products.*

Sample	Dosage (% a.i. or kg. a.i./ha.)	Formulation[a]	No. of applications	Days after last application	Residue (p.p.m.)[b]	Analytical method[c]	Country and year	Reference[d]
Coffee								
Whole beans	1.1 kg.	EC	2	0	1.1	GC	Puerto Rico (1968)	G.A.
				3	0.19	GC		
				7	0.14	GC		
				14	0.28	GC		
				28	0.09	GC		
Whole beans	1.1 kg.	EC	4	0	1.3	GC		
				3	0.37	GC		
				7	0.14	GC		
				14	0.08	GC		
Fermented beans	1.1 kg.	EC	4	39	<0.05	GC		
Whole beans	2.2 kg.	EC	2	0	3.7	GC		
				3	0.31	GC		
				7	0.19	GC		
				14	0.17	GC		
				28	0.11	GC		
Whole beans	2.2 kg.	EC	4	0	4.1	GC		
				3	0.6	GC		
				7	0.35	GC		
				14	0.31	GC		
Fermented beans	2.2 kg.	EC	4	39	<0.05	GC		
Olives								
Oil	0.03%	EC	2	62	0.8	GC	Italy (1966)	Eberle & Novak (1969)
Oil	0.06%	EC	2	62	1.8	GC		
Oil	0.05%	EC	1	86	0.3	GC		
Oil	0.05%	EC	2	50	0.8	GC		
Fruits	0.06%	EC	1	1	1.6	GC	Italy (1969)	G.B.
				9	0.39	GC		
				18	0.41	GC		
				35	0.18	GC		
				47	0.14	GC		
Tea								
Manufactured	0.03%	EC	1	21	0.03	GC	E. Pakistan (1968)	G.B.
			1	7	0.02	GC		
			3	21	0.04	GC		
Brewed tea	0.03%	EC	3	21	<0.002	GC		
Manufactured	0.07%	EC	1	7	0.03	GC		
			4	14	0.08	GC		
Brewed tea	0.07%	EC	4	14	<0.002	GC		
Tobacco								
Green leaves	0.6 kg.	EC	3	0	5.2	S	U.S.A. (1964)	G.A.
				7	2.1	S		
				14	0.26	S		
				22	0.17	S		
				30	0.08	S		
Cured leaves	0.6 kg.	EC	3	7	2.89	S		
Sand leaves	8 kg.	G	1	64	<0.01	GC	Switzerland (1968)	G.B.
Sand leaves	12 kg.	G	1	64	<0.01	GC		
Sand leaves	8 kg.	G	1	125	<0.01	GC		G.B.
Sand leaves	12 kg.	G	1	125	<0.01	GC		

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[a] WP = wettable powder, EC = emulsive concentrate, and G = granular formulation.

[b] Single star = average of two analyses and double star = average of three analyses.

[c] GC = gas chromatographic method, S = sulfide method, Sp = spectrophotometric method, Ph = phosphorus method, and ChI = cholinesterase inhibition method (see preceding review, this volume).

[d] G.A. = Geigy Chemical Corporation, Ardsley, New York, U.S.A.; G.B. = J. R. Geigy S. A., Basle, Switzerland; and M.-B. = personal communication from Professor H. Maier-Bode, Institute of Pharmacology, University of Bonn, G.F.R.

As the penetration of diazinon into treated plant parts is generally rather poor, residues and especially polar metabolites can be removed quite substantially by washing operations without detergents. From lettuce, 43 and 60 percent of the diazinon present were removed by washing three and nine times, respectively, in the household; a low residue of 0.36 p.p.m. on the lettuce could not be reduced by repeated washing (STOBWASSER 1961).

Residues on ready-to-market celery were effectively reduced by washing and trimming (FRANK and BRAUN 1969). One washing of tomatoes removed 88 percent of the residues. While cold washing was ineffective, residues in spinach could be reduced up to 60 percent by hot water blanching (FARROW *et al.* 1969). Under simulated commercial washing operations a spray rinse did not significantly reduce residues in spinach (RALLS *et al.* 1967). In the same study, washing with detergent lowered the levels of diazinon, diazoxon, and "pyrimidinol" from 1.8, 0.34, and 2.5 p.p.m. to 0.77, 0.18, and 2.0 p.p.m., respectively; steam blanching gave a general reduction to about 30 percent for diazinon and diazoxon but none for "pyrimidinol" and water blanching alone was even more effective.

Peeling removes most residues from fruits and root vegetables. Thus, no diazinon was determined in peeled canned carrots (STATHOPOULOS *et al.* 1971), nor in canned spinach which had been treated with diazinon five days before (see Table III).

The effect of heat processing and storage on the degradation of added pesticides was reported by ELKINS *et al.* (1971). After one year's storage at an ambient temperature of 100°F, diazinon added to canned spinach and apricots was completely degraded.

During wine-making two p.p.m. of diazinon added to grapejuice disappeared completely and displayed no noticeable effect on the fermentation process (PAINTER and KILGORE 1963).

IV. Diazinon residues in food moving in commerce and in the diet at the time of consumption

A significant reduction of residues occurs during food processing and preparation; thus diazinon is rarely detected in market-basket samples or in the total diet at the time of consumption.

Between 1964 and 1968, 2,396 market samples of fruits and vegetables produced in or imported to Sweden were analyzed for residues of organophosphorus pesticides by gas and thin-layer chromatography (RENVALL and ÅKERBLOM 1971 a); diazinon was found in only 0.8 percent of such samples, 75 percent of these containing residues between 0.01 to 0.09 p.p.m. Residues exceeding the 0.1 p.p.m. level were found in carrots (one sample) and lettuce (two samples).

In eight of 50 samples of olive oil collected on the Swedish market from 1965 to 1967 and analyzed for organophosphorus compounds

Table VIII. *Residues of diazinon in total-diet samples collected in the United States.*

Period of examination	No. of composites containing diazinon	Maximum diazinon levels detected (p.p.m.)	Reference
1965–1966	9 out of 216	0.051	DUGGAN *et al.* (1967)
1966–1967	2 out of 360	0.003	MARTIN & DUGGAN (1968)
1967–1968	1 out of 360	0.003	CORNELIUSSEN (1969)
1968–1969	14 out of 360	0.010	CORNELIUSSEN (1970)
1969–1970	21 out of 360	0.030	CORNELIUSSEN (1972)

(RENVALL and ÅKERBLOM 1971 b), diazinon was detected in concentrations ranging from 0.01 to 0.13 p.p.m.

Ready-to-eat foods were analyzed in the United States from June 1963 to April 1970 by the U.S. *Food and Drug Administration* before and after food preparation but only rarely was diazinon found. It was detected for the first time during the period June 1964 to April 1965. The frequency of diazinon residues in total-diet samples is given in Table VIII.

On the basis of such data daily pesticide intake figures were calculated (DUGGAN and WEATHERWAX 1967, DUGGAN and LIPSCOMB 1969, DUGGAN and CORNELIUSSEN 1972); the data for diazinon are reproduced in Table IX.

In these investigations a food intake almost twice that of the average food intake of the "average" individual was used. The data are thus considered the maximum possible dietary intakes of diazinon from

Table IX. *Average daily dietary intake of diazinon in the United States from 1966 to 1970.*

Period	Positive composites (%)	Daily dietary intake of diazinon	
		mg.	mg./kg. body weight[a]
1966	3	0.001	0.000020
1967	0.3	<0.001	0.000001
1968	0.3	<0.001	0.000001
1969	3.9	<0.001	0.000004
1970	5.8	0.001	0.000010
6-year average	—	—	0.000005

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[a] Based on an average body weight of 69.1 kg.

a well-balanced diet. The average value in the six-year period of 0.000005 mg./kg. body weight would imply a safety factor of 400 with regard to the acceptable daily intake of 0.002 mg./kg./day established by the World Health Organization.

V. Diazinon in soil

Contamination of the soil occurs either by direct application (against soil pests) or by run-off from treated plants.

After treatments according to good agricultural practice, initial residues averaged three p.p.m. Rapid loss of diazinon during the first weeks was followed by a much slower decline (GETZIN and ROSEFIELD 1966, READ 1969, BRO-RASMUSSEN *et al.* 1970). Approximately 50 percent of the diazinon applied disappeared two to four weeks after application (GETZIN and ROSEFIELD 1966, HARRIS 1969, BRO-RASMUSSEN *et al.* 1968 and 1970, MUNSON 1971) and more than 96 percent was lost after two to three months (*Geigy Chem. Corp.* 1968, MAIER-BODE 1968–1971, *Chesterford Park Research Station* 1964).

Dissipation and leaching studies conducted under field conditions showed that 50 percent of the diazinon disappeared from the soil two weeks after application. Of the total amount found, approximately 95 percent was in the zero-to-four-inch soil layer and the rest at depths between four and eight inches. Diazinon did not leach into lower soil zones but remained where applied (MUNSON 1971). In laboratory studies (*CIBA-GEIGY* 1972 c), the adsorption constants of diazinon ranged from 3.7 to 113.5 μg./g. in different soils; they rose as the organic matter content of the soil increased. Based on the adsorption constants and in comparison with the leaching behaviour of monuron, which is known to be a moderate "leacher" in the field, it can be assumed that the use of diazinon does not lead to a contamination of groundwater.

Though diazinon is one of the most widely and frequently used insecticides in sweet potatoes and onions in the United States, no residues in sweet potato fields (SAND *et al.* 1972) and only small average residues of 0.09 p.p.m. in about 20 percent of the onion fields were found (WIERSMA *et al.* 1972).

1. Degradation. — Degradation was mainly related to the soil type. After granular application of 1.8 lb. a.i./acre to a mineral soil (1.9 percent organic matter) and to a fen soil (17.1 percent organic matter) half of the diazinon applied had disappeared two and five weeks later, respectively. Seven months later, one and ten percent of the dose applied was determined in the mineral and fen soils, respectively (SUETT 1971). Soil organic matter thus seemed to protect diazinon from rapid degradation. In laboratory experiments the degradation of diazinon was also found to be related to temperature, moisture, and pH-value (MARGOT and GYSIN 1957, GETZIN 1968, KONRAD 1969, BRO-RASMUSSEN *et al.* 1968, GUNNER and ZUCKERMAN 1968, HARRIS 1971); higher temperatures, higher moisture levels, and increasing soil acidity accelerated the nonbiological degradation of diazinon. According to the experiments with steam-sterilized soil samples, BRO-RASMUSSEN *et al.* (1968) claimed that an essential part of the degradation was due to the activity of microorganisms.

GUNNER *et al.* (1966) isolated a bacterium from the soil which utilized the diazinon molecule as the sole source of sulfur, phosphorus, carbon, and nitrogen, but the practical importance of this organism as a contributor to diazinon degradation was not determined. GUNNER and ZUCKERMAN (1968) reported that two microorganisms, an *Arthrobacter* sp. and a *Streptomyces* sp., occurring as the predominant successive microflora in soils treated with diazinon, were able to degrade the pyrimidine ring to carbon dioxide.

In a flooded soil (pH 6.6) which had previously been treated with diazinon, the insecticide persisted for only ten to 15 days, but when the compound was applied to the flooded soil for the first time the persistence was about two months (SETHUNATHAN 1971). Loss of diazinon from sterilized samples of a neutral and an alkaline soil was much slower than from sterilized samples of the same soils, thus indicating microbial participation in degradation (SETHUNATHAN and MACRAE 1969). In fact, repeated applications of diazinon resulted in the build-up of a special microflora that had been found to accelerate the degradation of the compound (SETHUNATHAN *et al.* 1971, SETHUNATHAN 1971 b).

LICHTENSTEIN (1966) and LICHTENSTEIN *et al.* (1968) investigated the degradation of diazinon after addition of detergents to the soil after autoclaving and sterilizing with sodium azide. Surprisingly, the addition of high amounts of detergents increased the persistence of diazinon, although the number of bacteria was ten to 50 times higher than without detergents. Also unexpected was the finding that diazinon was no longer detectable in the soil after a two-week incubation with sodium azide although the bacteria were strongly reduced by this sterilizing agent. The effect of sodium azide is not exclusively a sterilizing one, but it was observed that it catalyzed the hydrolysis of diazinon in distilled water.

2. Metabolites. — In experiments with ^{14}C-ring labelled diazinon, GETZIN (1967) found that 35 percent of the ^{14}C was evolved as $^{14}CO_2$ after 20 weeks. Little $^{14}CO_2$ was released from soil fumigated with propylene oxide in which the microbial activity was reduced. However, greater amounts of "pyrimidinol" were recovered from soil fumigated as compared to the nonfumigated soil. Hence, the soil microflora apparently play a major role in the degradation of the breakdown products, but not in the initial hydrolysis of the parent molecule. The oxygen analog, diazoxon, was not detected in soils treated with diazinon.

In flooded soils hydrolysis was also a major step in degradation, leading to "pyrimidinol" which has no anticholinesterase activity (MARGOT and GYSIN 1957). This hydrolysis product tended to accumulate temporarily under submerged conditions where the bulk of the soil microflora was anaerobic and oxidation was slow (SETHUNATHAN and YOSHIDA 1969). "Pyrimidinol" was found to be further degraded to "hydroxy-pyrimidinol and to carbon dioxide (LAANIO *et al.* 1972), and drying of the soil or increasing aeration during seed-bed preparation seemed to speed up the process (SETHUNATHAN and YOSHIDA 1969).

VI. Diazinon in water

GOMAA *et al.* (1969) have studied the degradation of diazinon and diazoxon in water under varying conditions of temperature and pH. Both compounds were quantitatively hydrolyzed to "pyrimidinol" and diethylphosphoric acid. The half-life of diazinon in water with different pH values was as follows: hydrolysis was extremely rapid under acidic conditions (pH 3.1, approximately 12 hours), whereas in neutral or slightly alkaline solutions the rates were lower (pH 7.4 and 9.0, 184 and 136 days, respectively). Raising the pH from 9.0 to 10.4 accelerated the degradation process appreciably (pH 10.4, 24 days). According to COWART *et al.* (1971), diazinon hydrolyzed within two weeks to more than 50 percent in a slightly acidic water (pH 6). At 20°C diazoxon hydrolyzed much faster than diazinon under comparable conditions. Under acidic conditions diazoxon hydrolyzed 30 times faster than the parent compound (GOMAA *et al.* 1969).

In water from a rice field which had been treated previously with diazinon, the insecticide was degraded within three to five days; the hydrolysis to "pyrimidinol" was complete within about 75 hours and in the next 25 hours this product was completely degraded to carbon dioxide. During the same period, degradation in water from a field not previously treated was negligible. Several bacteria, especially a coccoid bacterium, isolated from water of a diazinon-treated field degraded the compound in a pure culture. Actinomycetes also seemed to be involved, because streptomycin prevented the breakdown of diazinon. The microbial flora induced by diazinon were not able to degrade other related insecticides (HIRANO and YUSHIMA 1969, SETHUNATHAN 1971 a and b, SETHUNATHAN and PATHAK 1972).

Summary

In this review are compiled the available residue data on diazinon used at recommended rates in fruits, vegetables, cereals, and forage crops as well as in soil and water. The data originate from field trials in various countries. The rate and the nature of degradation

of the insecticide on plants and in soil and water are described briefly.

On fruits, grapes, and citrus fruits, residues of one to five p.p.m. are found immediately after treatment; on the average they decrease by 50 percent in four days.

On leafy vegetables, hops and forage crops (alfalfa, grass) higher initial residues but also shorter half-life periods are measured.

Residues on other vegetables (beans, tomatoes, cucumbers), cereal products, sugarcane, tea, and coffee are low (<0.5 p.p.m. seven days after treatment).

In crops with a high content of oil, such as carrots and oil seed plants, degradation of diazinon seems retarded.

An average of three p.p.m. of diazinon is found in the soil immediately after treatment. The half-life period is two to four weeks. The rate of degradation is influenced by the nature of the soil and the climatic conditions.

Diazinon is only slightly translocated in plants. It is degraded mainly by hydrolysis of the thiophosphorus ester and by subsequent side-chain hydroxylation of the resulting 2-isopropyl-4-methyl-6-hydroxyprimidine. This hydrolysis product was detected in peas, beans, spinach, rice, and tomatoes. Only traces of diazoxon, the oxygen analog of diazinon, are ascertained in pome fruits, vegetables, rice, and olive oil. Equally low are the residues of "hydroxy-diazinon" in cabbage, beans, and rice.

The degradation of diazinon in soil as well as in water of flooded rice fields also occurs through hydrolysis of the thiophosphorous ester with ensuing oxidation to carbon dioxide. The rate is considerably enhanced by an increase in the acidity of the water as well as by an adaptation of the microflora through preceding treatment with the insecticide.

Storage and processing (washing, peeling, cooking, and canning) of treated fruit and vegetables considerably reduce residues remaining at harvest.

Studies in the United States showed the maximum possible intake of diazinon with the daily human diet to be 0.000005 mg./kg.

Résumé*

Diazinon. II. Résidus dans les plantes, le sol, et l'eau

Dans ce chapitre sont présentées des valeurs résiduelles du diazinon, déterminées après des traitements aux dosages recommandés, sur les arbres fruitiers, les légumes, les céréales et dans les cultures

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* Traduit par G. DUPUIS et N. TAMINI.

fourragères ainsi que dans le sol et l'eau. Ces valeurs proviennent d'essai de plein champ effectués dans divers pays. On a également mentionné le comportement et la dégradation de l'insecticide dans les plantes, dans le sol et dans l'eau.

Aussitôt après un traitement, on a trouvé 1–5 p.p.m. sur les fruits, des raisins et des agrumes; ces résidus diminuent de 50 pour cent au bout de quatre jours en moyenne.

Sur les légumes feuillus, du houblon et des cultures fourragères (luzerne, graminées), on a noté des valeurs résiduelles initiales plus élevées, mais aussi des temps de demi-dégradation plus courts.

Quant aux résidus trouvés sur d'autres légumes (haricots, tomates, concombres), des céréales, de la canne à sucre, du thé et du café, ils sont faibles (<0.5 p.p.m. sept jours après le traitement).

Dans les cultures avec une concentration élevée en huile, comme les carottes et les plantes aux graines riches en huile, la dégradation du diazinon se trouve retardée.

Après un traitement direct on enregistre en moyenne dans le sol trois p.p.m. de diazinon. Le temps de demi-dégradation atteint deux à quatre semaines. La rapidité de la dégradation dépend du type de sol et des conditions climatiques.

Le diazinon appliqué sur la plante subit une translocation partielle et une dégradation, principalement par hydrolyse de l'ester thiophosphorique qui donne naissance au "pyrimidinol," suivie d'une hydroxylation de la chaîne latérale avec formation de 2-(1′-hydroxy-1′-méthyl) éthyl-4-méthyl-6-hydroxy-pyrimidine. Parmi les métabolites, il faut mentionner surtout le "pyrimidinol" qui a été décelé, entre autres, dans les pois, les haricots, les épinards, les tomates, le riz etc. Le diazoxon, l'analogue oxygéné du diazinon, est un métabolite transitoire dont on n'a pu déterminer que des traces dans les fruits à pépins, les légumes, le riz et l'huile d'olive. De même, des résidus d'hydroxy-diazinon, thiophosphate de O,O-diéthyl-O-[2(1′-hydroxy-1′-methyl)éthyl-4-méthyl-pyrimidine-6-yl] n'ont été décelés qu'en faibles quantités dans les choux, les haricots et le riz.

La dégradation du diazinon dans le sol et dans l'eau (rizière immergées) se produit par hydrolyse de l'ester phosphorique et oxydation consécutive en CO_2. La vitesse de dégradation est nettement influencée par l'acidité de l'eau, ainsi que par une microflore bien déterminée, qui se forme seulement après un traitement précédent au diazinon.

La préparation des aliments (lavage, cuisson, mise en conserve) et la conservation des fruits et des légumes triatés réduisent encore de manière déterminante les quantités de résidus subsistant lors de la récolte.

Selon des expériences faites aux Etats-Unis, l'absorption possible de diazinon dans la nourriture quotidienne ne dépasse pas 0,000005 mg./kg.

Zusammenfassung*

Diazinon. II. Rückstände in Pflanzen, im Boden, und im Wasser

Im vorliegenden Kapitel sind die Rückstandswerte von Diazinon zusammengestellt, wie sie nach Behandlungen in empfohlenen Dosierungen auf Obst-, Gemüse-, Getreide-und Futterkulturen sowie im Boden und Wasser vorkommen. Die Daten stammen von Feldversuchen aus verschiedenen Ländern. Das Verhalten und der Abbau des Insektizids auf pflanzlichen Unterlagen, im Boden und im Wasser werden kurz beschrieben.

Auf Obst, Trauben und Citrusfrüchten werden unmittelbar nach einer Behandlung Rückstände von 1–5 p.p.m. nachgewiesen; sie verringern sich um 50 Prozent in durchschnittlich vier Tagen.

Auf Blattgemüsen, Hopfen und Futterkulturen (Luzerne, Gras) werden höhere Anfangsrückstände aber auch kürzere Halbwertzeiten gemessen.

Gering sind die Rückstände auf anderen Gemüsen (Bohnen, Tomaten, Gurken), Getreideprodukten, Zuckerrohr, Tee und Kaffee ($<$0,5 p.p.m. sieben Tage nach der Behandlung).

In Kulturen mit einem hohen Ölgehalt, wie Karotten und Ölsaatpflanzen, wird der Abbau von Diazinon verzögert.

Durchschnittlich drei p.p.m. Diazinon finden sich im Boden nach Diretkbehandlung unter Praxisbedingungen. Die Halbwertzeit beträgt zwei bis vier Wochen. Die Abbaugeschwindigkeit hängt ab vom Bodentyp und den klimatischen Bedingungen.

Das auf die Pflanze applizierte Daizinon wird nur teilweise transloziert. Es wird hauptsächlich durch Hydrolyse des Thiophosphorsäureesters und nachfolgender Seitenketten-Hydroxilierung des enstandenen Pyrimidinols (2-Iso-propyl-4-methyl-6-hydroxy-primidine) abgebaut. Dieses Hydrolyseprodukt ist in Erbsen, Bohnen, Spinat, Reis und Tomaten nachgewiesen worden. Das Sauerstoffanaloge, Diazoxon, ist ein transitorischer Metabolit, der lediglich in Spuren bei Kernobst, Gemüse, Reis und in Olivenölfestgestellt werden konnte. Ebenso gering sind die Rückstände von "Hydroxy-diazinon," *O,O*-Diethyl-*O*-(2-(1′-hydroxy-1′-methyl)ethyl-4-methyl-pyrimidin-6-yl)-phosphorothioate, auf Kohl, Bohnen und Reis.

Der Abbau von Diazinon im Boden und im Wasser (überflutete Reisfelder) geschieht hauptsächlich durch Hydrolyse des Phosphorsäureesters mit nachfolgender Oxidation des Heterozyclus zu CO_2. Die Abbaugeschwindigkeit wird maßgeblich beeinflußt durch die

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* Übersetzt von der Autorin.

Azidität des Wassers sowie durch eine bestimmte Mikroflora, die sich nach vorhergehenden Diazinonbehandlungen stark vermehrt hat.

Verarbeitung (Waschen, Schälen, Kochen, Einmachen) und Konservierung von behandeltem Obst und Gemüse reduzieren die noch bei der Ernte verbliebenen Rückstände beträchtlich.

Nach Untersuchungen in den Vereinigten Staaten wird die mögliche, maximale Aufnahme von Diazinon mit der täglichen Nahrung mit 0,000005 mg./kg. angegeben.

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