Fate of Pentachlorophenol-^{14}C in Rice Plants under Controlled Conditions

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Pentachlorophenol-^{14}C was applied to soil (23 kg/ha), and rice plants were grown over two vegetation periods under flooded conditions. The uptake of radioactivity by plants was 12.9% in the first and 2.5% in the second year. Rice roots (first year) contained 0.14% of the applied radioactivity as free pentachlorophenol; 0.06% was conjugated pentachlorophenol, 0.43% free and conjugated lower chlorinated phenols, 0.07% anisoles, 0.01% dimethoxytetrachlorobenzenes, 0.03% 1,2-dihydroxy- and/or monohydroxymonomethoxytetrachlorobenzene, 0.48% polar nonhydrolyzable substances, and 3.95% unextractable residues. In the straw (first year), neither pentachlorophenol nor lower chlorinated phenols or anisoles were detected; the main metabolite (0.63%) was probably tetrachlorobenzoquinone. In the grains, low radioactive residues (0.12% of applied radioactivity or 4 ppm equivalent to pentachlorophenol) could not be identified. In the second year (pentachlorophenol residues in soil corresponding to ~8.4 kg/ha), the portion of unextractable residues in the plants increased, and lower chlorinated conjugated phenols were identified.

Pentachlorophenol (PCP) and its salts are used as fungicides, herbicides, insecticides, and molluscicides in agriculture. They have been used in Japan in large quantities as herbicides in rice fields. Since 1971 their use is restricted because of their high fish toxicity (Kuwatsuka and Igarashi, 1975). Today PCP is used mainly as a wood preservative, which has led to its widespread presence in the environment.

A number of PCP metabolites have been isolated from soil, microorganisms, aquatic systems, animals, and abiotic systems (Munakata and Kuwahara, 1969; Tashiro et al., 1970; Suzuki and Nose, 1971; Chu and Kirsch, 1972; Ide et al., 1972; Kuwatsuka and Igarashi, 1975; Kobayashi, 1977; Ahlborg, 1977; Wong and Crosby, 1977; Murthy et al., 1979; Rott et al., 1979). Information about PCP residues in rice (Haque et al., 1978) and maize (Lu et al., 1977) is available but little is known about its degradation products. Thus, it was thought of interest to study its metabolism in plants. In this paper metabolism of PCP in rice plants is reported.

MATERIALS AND METHODS

Apparatus. The radioactivity in various extracts was measured in a liquid scintillation counter (Packard Tricarb, Model 3375 or 3380) with external standardization. Unextractable residues were determined by automatic combustion (Oxymat, Intertechnique, or Packard Oxidizer B 306). Thin-layer chromatography plates were scanned for radioactive substances on a scanner supplied by Berthold/Frieseke GmbH, Karlsruhe, Federal Republic of Germany. Gas–liquid chromatographic (GLC) analysis was performed on a Carlo Erba unit with an EC detector, fitted with a glass capillary column 50 m long coated with methyl silicone (SP 2100) (Carbowax 20M deactivated) and with a Hewlett-Packard integrator, Model 3388 A. The following conditions were used: temperature 135–220 °C, programmed at 1.5 °C/min; detector temperature 250 °C; carrier gas N_2 (0.26 mL/min). A gas chromatograph–mass spectrometer (GC–MS) LKB 9000, from LKB-Produkter, Bromma, Sweden, was used for mass spectrometry.

Reagents. PCP-^{14}C was synthesized in our laboratory (Sandrock et al., 1978). Its purity was determined by thin-layer chromatography (solvent: petroleum ether–benzene–acetic acid, 2:2:1) and was found to be >99%.

A scintillation liquid based on dioxane was used for assaying extracts and TLC zones. A toluene-based scintillator containing phenethylamine was used for assaying $^{14}CO_2$ from samples combusted in the Oxymat. For the oxidizer a Packard basic scintillator was used. Silica gel G (Macheray, Nagel u. Co.) was used for the preparation of TLC plates. Ready-made silica gel plates (Merck; 0.25 and 2 mm) and Al_2O_3 plates (Type E, aluminum oxide 60; 0.25 mm) were also used. For column chromatography, silica gel 60 (0.063–0.200 mm; 70–230-mesh ASTM; Merck) and Sephadex LH-20 (25–100 μm; Pharmacia Fine Chemicals) were used.

Authentic Samples. PCP was purchased from Riedel-DeHaen AG, Selze-Hannover, purity 99% (checked by TLC using as the solvent petroleum ether–benzene–acetic acid, 2:2:1). All three tetrachlorophenol isomers were purchased from Fluka AG, Buchs SG. 2,3,4-, 2,3,6-, and 3,4,5-trichlorophenols were from EGA Chemie KG. 2,3,5-Trichlorophenol was from Aldrich Chemical Co., Inc. 2,4,5-Trichlorophenol was from Merck-Schuchardt. 2,4,6-Trichlorophenol was from Hoechst.

All three tetrachloroanisole isomers, all six trichloroanisole isomers, and pentachloroanisole were synthesized by methylation of respective phenols with diazomethane. 1,2-Dimethoxytetrachlorobenzene and 1,4-dimethoxytetrachlorobenzene were prepared by methylation of tetrachlorocatechol and tetrachlorohydroquinone with $(CH_3)_2SO_4$ in methanolic KOH. For methylation of metabolites and authentic samples, diazomethane was freshly prepared from (*p*-tolylsulfonyl)methylnitrosamide and KOH in diethyl ether and then distilled.

Application and Cultural Conditions. A Kick-Brauckmann vessel (Kick and Grosse-Brauckmann, 1961) and a 7-L plastic vessel were filled with 8 and 7 kg of sandy soil, respectively. In order to maintain adsorption properties of soil and bioavailability of PCP to plants as natural as possible, and in order to include also natural soil metabolites into the investigation, we did not sterilize the soil. Before seeding the soil was treated with fertilizer corresponding to 1.6 g of N, 2.4 g of K_2O, and 1.2 g of P_2O_5. Rice plants (*Oryza sativa* var. *aristata*) were allowed to grow in a phytotron (Vötsch) under flooded conditions. Table I reports the summary of the climate conditions as well as the analysis of soil. When the plants were 3 months old, 171.7 mg of PCP-^{14}C, specific activity 0.25 mCi/mmol, were dissolved in a mixture of 3 mL of diethyl ether and 37 mL of acetone and applied dropwise onto the soil of the two

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Table I. Cultural Details for Rice Plants Grown with Pentachlorophenol-^{14}C

soil analysis	climate	1st and 2nd year	application of PCP-^{14}C	
clay, 7.4%	day temperature, °C	24	rate of application, kg/ha	23 (85.86 mg)
silt, 7.0%	night temperature, °C	18	specific activity, mCi/mmol	0.25
sand, 85.6%	photoperiod hours	16		
humus, 1.1%	light intensity, klx	22		
pH 5.7	air humidity, % (max)	80		
	air humidity, % (min)	65		

Table II. Uptake of Radioactivity into Rice Plants Grown in Soil Treated with Pentachlorophenol-^{14}C (Percent of Applied Radioactivity)

growing period	plant part	extractable radioact	unextractable radioact	total radioact
first year[a]	roots	1.22	3.95	5.17
	straw	3.14	4.37	7.51
	husks	0.02	0.03	0.05
	grain	0.02	0.10	0.12
	total plants	4.40	8.45	12.85
second year[b]	roots	0.25	1.57	1.82
	tops	0.36	0.30	0.66
	total plants	0.61	1.87	2.48

[a] Mean of two experiments. [b] One experiment.

pots (20 mL in each pot). This dosage corresponded to 23 kg/ha, as used in agricultural practice for rice fields. The relatively low specific activity of the PCP-^{14}C used was necessary since the use of higher radioactivities is not possible in open containers in the laboratory. After 7 months and 16 days the plants were harvested. The soil in the Kick-Brauckmann vessel was stored at −20 °C for 5 months and then used for rice growing for a second vegetation period. The initial concentration in the second vegetation period corresponded to 8.4 kg/ha. The soil was fertilized with 10 g of "Nitrophoska blau" and sown with 43 seeds of rice. After 5 weeks the soil was flooded and the plants were grown in a phytotron under the conditions given in Table I.

Preparation of Plant Material for Analysis. The rice plants of the first vegetation period were separated into roots, straw, husks, and grain. Rice plants of the second vegetation period were separated only into roots and tops. The plants were homogenized and extracted with methanol for 48 h in a Soxhlet apparatus. Radioactivity in the extracts was counted by a liquid scintillation counter. The unextractable ^{14}C in various samples was measured by liquid scintillation after oxidation to $^{14}CO_2$ (Table II).

The unextractable residues bound to cellulose and/or lignin of the plant were determined by the method reported in Honeycutt and Adler (1975). For analysis of soil, see Weiss et al. (1982). No detectable radioactivity was left at the walls of the containers at the end of the experiments.

Isolation and Identification of the Conversion Products. For isolation of metabolites, all the methanol extracts were evaporated to dryness in a rotary evaporator (condenser −20 °C) and then subjected to liquid–liquid partition with a diethyl ether–water system at different pHs (Figure 1). By this procedure, separation into neutral, organic-soluble substances (fraction A), acidic (fraction B) substances, and water-soluble substances (fraction C) was achieved; however, a very small portion of the phenolic group was found also in fraction A and a very small portion of anisoles also in fraction B (see Tables III–V).

Isolation of Metabolites from Roots, First Vegetation Period (Table III). After concentration of the etheric *fraction A* of the root extract of the first year on a rotary evaporator, the fraction was separated into radioactive zones on preparative silica gel plates (20 × 20 cm; 2 mm) by using chloroform–cyclohexane–acetic acid (9:9:2). Three zones were obtained: a very polar zone A I (R_f 0, 0.1% of applied radioactivity), a zone A II (R_f 0.45, 0.02%), and a less polar zone A III (R_f 0.78, 0.08%).

Table III. Identification of Extractable Radioactive Residues in Roots of Rice Grown in Soil Treated with Pentachlorophenol-^{14}C (First Growing Period; in Percent of Applied Radioactivity)

radioact zone (TLC)	R_f (TLC)	solvent system (TLC)	%	identified products
A III/1	0.25	benzene	0.01	2,3,4,5-tetrachlorophenol plus pentachlorophenol
A III/2.1.1	0.35	20% benzene in pentane	<0.01	3,4,5-trichloroanisole
A III/2.1.2	0.77	20% benzene in pentane	0.01	1,2- plus 1,4-dimethoxytetrachlorobenzene
A III/2.2	0.26	hexane	0.02	2,3,4,5-tetrachloroanisole
A III/2.3	0.46	hexane	0.04	pentachloroanisole
A II	0.17	benzene	0.02	2,3,5-trichlorophenol
A I	0	chloroform–cyclohexane–acetic acid (9:9:2)	0.10	0.02% (appl radioact) conjugated 3,4,5-trichlorophenol
B2/I	0.26	hexane	<0.01	2,3,4,5-tetrachloroanisole
B2/II	0.45	hexane	<0.01	pentachloroanisole
B3/I	0.20[a]	hexane	0.04	2,3,4,5-tetrachlorophenol
B3/II	0.40[a]	hexane	0.01	pentachlorophenol
B4/I	0.11[a]	hexane	0.03	1,2-dihydroxy- and/or monohydroxy monomethoxy-tetrachlorobenzene
B4/II 1	0.43[a]	pentane	0.29	3,4,5-trichlorophenol and pentachlorophenol
B4/II 2	0.54[a]	pentane	0.08	pentachlorophenol
B5–8	0	chloroform–cyclohexane–acetic acid (9:9:2)	0.30	0.14% (appl radioact) conjugated 3,4,5-trichlorophenol and pentachlorophenol
C, nonhydrolyzable part			0.24	not identified
C, hydrolysate	0.84[a]	benzene	0.03	conjugated 3,4,5-trichloro- and 2,3,4,5-tetrachlorophenol

[a] R_f values given after methylation with CH_2N_2.

Table IV. Characterization of Extractable Radioactive Residues in Straw of Rice Grown in Soil Treated with Pentachlorophenol-^{14}C (First Growing Period; in Percent of Applied Radioactivity)

radioact zone	R_f	solvent system	%	identified products
A I/1	0	benzene-ethyl acetate-acetone (9:9:2)	0.06	highly polar, not hydrolyzable
A I/2	0.21	benzene-ethyl acetate-acetone (9:9:2)	0.27	polar, not hydrolyzable, cannot be methylated
A I/3	0.42	benzene-ethyl acetate-acetone (9:9:2)	0.10	polar, not hydrolyzable, cannot be methylated
A I/4	0.60	benzene-ethyl acetate-acetone (9:9:2)	0.04	polar, not hydrolyzable, cannot be methylated
A II	0.54	chloroform-cyclohexane-acetic acid (9:9:2)	0.63	probably tetrachlorobenzoquinone
A III	0.6-1.0	chloroform-cyclohexane-acetic acid (9:9:2)	0.04	not identified
B I	0	chloroform-cyclohexane-acetic acid (9:9:2)	0.45	very polar, not hydrolyzable
B I	0			
B II/1	0.30	hexane-ethyl acetate-acetone (12:7:1)	0.03	cannot be methylated, not identified
B II/2	0.60	hexane-ethyl acetate-acetone (12:7:1)	0.03	cannot be methylated, not identified
C, nonhydrolyzable part			1.47	not identified
C, hydrolysate	0.23[a]	benzene	0.02	not identified

[a] R_f value given after methylation with CH_2N_2.

Table V. Identification of Extractable Radioactive Residues in Roots and Straw of Rice Grown in Soil Treated with Pentachlorophenol-^{14}C (Second Growing Period; in Percent of Applied Radioactivity)

plant part	radioact fraction	R_f	solvent system	%	identified products
roots	nonhydrolyzable polar substances			0.07	not identified
roots	hydrolysate[a]	0.86[a]	benzene	0.02	conjugated trichloro-, tetrachloro-, and pentachlorophenol
straw	nonhydrolyzable polar substances			0.20	not identified
straw	hydrolyzate	0.85[a]	benzene	0.01	conjugated dichloro-, trichloro-, and tetrachlorophenol

[a] R_f values given after methylation with CH_2N_2.

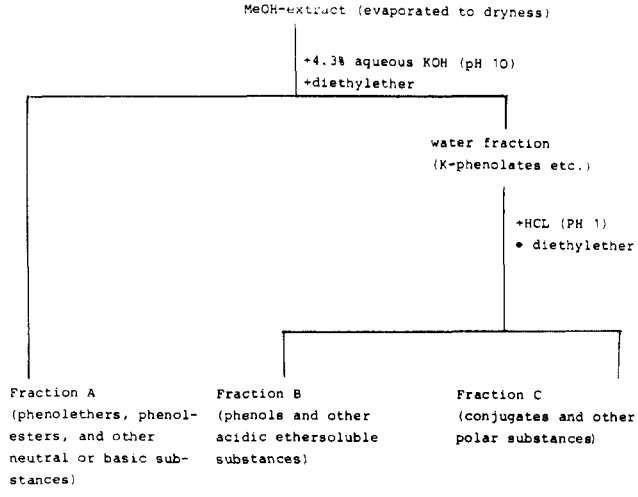

Figure 1. Fractionation of methanol extracts of plants by liquid–liquid partition at different pH values.

Zone A III on further TLC analysis with different solvent systems and purification was seen to be a mixture of radioactive substances that were then eluted from the TLC coating and subjected to GLC and GLC–MS analysis.

Zone A II after methylation with CH_2N_2 and repeated purification on TLC was seen to be a single substance that was also subjected to GLC and GLC–MS analysis.

Zone A I, a very polar zone, was hydrolyzed with 9 N H_2SO_4 at 70 °C for 8 h, diluted with water, and extracted with diethyl ether. The hydrolysate was methylated with freshly prepared CH_2N_2 and subjected to TLC analysis. It was observed that only a part of the radioactivity was hydrolyzed (0.02% on the basis of applied radioactivity); this was identified by GLC and GLC–MS analysis.

Fraction B after concentration was subjected to column chromatography (column 50 × 2.5 cm, silica gel 100 g, 0.063–0.2 mm), and the column was eluted with different solvents. The eluates containing sufficient amounts of radioactivity were separated into various radioactive substances on TLC plates. The percentages and R_f values are given in Table III. The nonpolar substances were identified directly by GLC and GLC–MS analysis, and a part of the polar fractions (eluates B5–8, Table III) was identified after hydrolysis and methylation.

Fraction C was hydrolyzed with 9 N H_2SO_4 at 70 °C for 8 h and extracted with diethyl ether. Only a very small part of this fraction was hydrolyzable. The hydrolysate was methylated and after purification on TLC plates was subjected to GLC and GLC–MS analysis.

Isolation of Metabolites from Rice Straw, First Vegetation Period (Table IV). Fractions A–C (Figure 1) of the rice straw extract after the first year were separated into various radioactive substances by column chromatography, TLC, and hydrolysis. The main metabolite of fraction A (0.63% of the applied radioactivity, A II in Table IV) was subjected to GLC–MS. The other radioactive substances were insufficient for instrumental analysis.

Isolation of Metabolites from Roots and Rice Straw, Second Vegetation Period (Table V). Isolation and identification of conversion products from roots and straw of the second year were carried out only for the polar fraction C (Figure 1) because identification of fractions A and B failed due to very small amounts of ^{14}C. Fraction C was analyzed as that of the first year. The extracts of rice grains and husks were not subjected to any analytical procedure because of insufficient radioactive material.

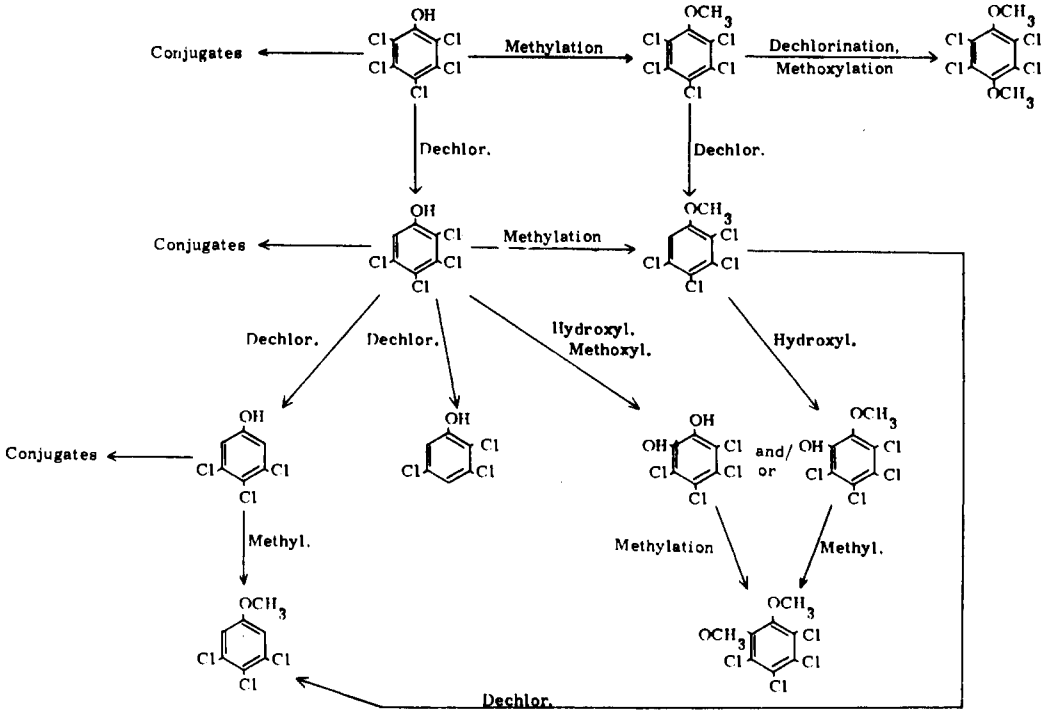

Figure 2. Conversion pathways of pentachlorophenol in rice roots.

Procedural Recoveries. The mean recovery for each chromatographic step was about 90%; losses of about 10% (evaporation and extraction losses, etc.) could not be avoided. No chemical alteration of the identified metabolites occurred, as evident from identical chromatographical behavior between the purification steps. Where many purification steps had to be performed, only microgram or nanogram amounts were left, which, however, were sufficient for mass spectrometric analysis.

RESULTS

Uptake of Radioactivity from Soil. In the first vegetation period, rice plants took up 12.9% and in the second vegetation period 2.5% of the total radioactivity (Table II). Thus, the uptake of radioactivity by the rice plants in the second vegetation period was less than in the first vegetation period. This is partly due to the smaller amount of available radioactivity for the plants in the soil that was, in the second year, only 36% of that of the first year, partly due to the differences in extractability of soil residues between the two years, and partly due to other, unknown factors (Weiss et al., 1982). Table II also demonstrates that the major part of the radioactivity in the plant parts is not extractable; i.e., it is bound to the macromolecules of natural plant constituents. The grains contained low residues (0.12% of applied radioactivity), corresponding to about 4 $\mu g/g$ (μg equivalent to pentachlorophenol). The ratio of unextractable to total residues increased in the second growing period.

Identification of Metabolites. Identification of metabolites was carried out with the root and straw extract of both years. The conversion products isolated from the rice roots of the first vegetation period are shown in Table III. The radioactivity in the grain and husk extract was too low for identification.

Fraction A (Figure 1) from the *root extract of the first year* on TLC analysis gave three radioactive zones (see Materials and Methods). Zone A III (Table III) upon further TLC analysis was found to be a mixture of two groups of radioactive substances. One group (the major one, A III/2 in Table III) was found to be a mixture of anisoles, and a minor group was found to be a mixture of free phenols. The major substance in the anisole group was identified by GLC and GLC–MS as pentachloroanisole. The retention time on GLC and the fragmentation pattern were identical with those of the authentic sample. The other substances identified in this group were 2,3,4,5-tetrachloroanisole, 3,4,5-trichloroanisole, and a mixture of 1,2- and 1,4-dimethoxytetrachlorobenzene. The identification of these compounds was done as in the case of pentachloroanisole. The free phenols in the minor group (A III/1 in Table III) were methylated with freshly prepared CH_2N_2. After TLC purification, the methylated products were subjected to GLC and GLC–MS analysis. The retention time on GLC and the mass spectrum were identical with those of 2,3,4,5-tetrachloroanisole and pentachloroanisole. Zone A II (Table III) after repeated TLC purification and methylation with CH_2N_2 was found to be a single compound. Its mass spectrum, retention time on GLC, and R_f on TLC were identical with those of 2,3,5-trichloroanisole. The polar fraction (zone A I, Table III) was a group of conjugates that, upon hydrolysis, was shown by GLC–MS analysis to contain, among other unidentified products, 3,4,5-trichlorophenol.

Fraction B (Figure 1) after subjection to various analytical procedures as described above was found to be a mixture of phenols and anisoles. The chemical nature and R_f values are given in Table III. The radioactive zone B4/I (Table III) after methylation was identified as 1,2-dimethoxytetrachlorobenzene. It is possible that the substance prior to methylation was a monomethoxymonohydroxytetrachlorobenzene or a dihydroxytetrachlorobenzene.

Fraction C (Figure 1), the polar one, was hydrolyzed, methylated with CH_2N_2, and shown to contain, among other unidentified products (Table III), 3,4,5-trichloroanisole and 2,3,4,5-tetrachloroanisole by GLC and GLC–MS.

The only metabolite identified from *rice straw of the first vegetation period* (A II in Table IV) in mass spectrometry had a characteristic isotope distribution pattern for four chlorine atoms and a mole peak at 244. It was

assumed to be a tetrachlorobenzoquinone. Detailed chemical analysis was not possible because of an inadequate amount. The other ^{14}C-labeled substances present in the extract were highly polar and were not identified.

Fraction C from *roots of the second year* (Table V) was analyzed the same way as reported above. The substances identified by GLC–MS after hydrolysis were trichlorophenol, tetrachlorophenol, and pentachlorophenol. The position of the chlorine atoms in the tri- and tetrachlorophenols was not established.

The *straw extract of the second year* (Table V) was found to contain conjugated di-, tri-, and tetrachlorophenols. The spectra were mixed because of large quantities of biological material associated with the metabolites. Comparison with authentic samples was not done because of insufficient amount of these metabolites.

The *unextractable residues* incorporated in lignin and/or cellulose of rice straw (first year) were determined by the method reported by Honeycutt and Adler (1975). In cellulose 0.5% and in lignin 0.8% of the applied radioactivity were incorporated. The nature of the incorporated activity is not known.

DISCUSSION

The analyses carried out show that the major part of the ^{14}C taken up from pentachlorophenol-^{14}C-treated soil was bound to the plant material. The major part of the extractable radioactivity, also, was neither hydrolyzable nor identifiable. The identifiable portion of radioactive substances in the rice roots of the first year consisted of only 0.14% of free unchanged pentachlorophenol and 0.06% of conjugated pentachlorophenol (percent based on added PCP-^{14}C). Free and conjugated lower chlorinated phenols (0.43%) and anisoles (0.07%) demonstrate that the degradation of pentachlorophenol in rice roots includes dechlorination and methylation pathways, resulting in one of the three possible tetrachlorophenols, in one of the three existing tetrachloroanisoles, in two of the six existing trichlorophenols, and in one of the six existing trichloroanisoles (besides pentachloroanisole). The presence of 0.01% of two dimethoxytetrachlorobenzenes and 0.03% of a 1,2-dihydroxy- and/or monohydroxymonomethoxytetrachlorobenzene shows the participation of oxidative processes. Figure 2 presents the metabolic pathways concluded from the metabolites identified.

Since the plants grew in normal, nonsterilized soil, the question whether all of these metabolites have been formed in the plants or whether part of them have been formed by soil microorganisms and taken up by plants cannot be completely answered. However, since up to now no oxidation products of pentachlorophenol have been detected in soil, at least the dimethoxytetrachlorobenzenes, the dihydroxytetrachlorobenzene, and/or the monohydroxymonomethoxytetrachlorobenzene may be regarded as real plant metabolites. For the conversion products in soil, see Weiss et al. (1982).

In the rice straw of the first year, only one metabolite could be chemically characterized. Its chemical nature (probably tetrachlorobenzoquinone) and the presence of numerous unidentifiable and unstable products suggest an oxidative degradation pathway via quinoid structures, resulting also in unextractable residues. The metabolites of the second year indicate continuing dechlorination (elimination of up to three chlorine atoms) and continuing formation of unextractable residues.

It may be concluded from these experiments that pentachlorophenol is not persistent in rice plants but is degraded and incorporated mostly into natural plant constituents within one growing period. However, since the chemical forms of binding are not known, the toxicological consequences of these bound residues cannot yet be assessed.

LITERATURE CITED

Ahlborg, U. G. In "Pentachlorophenol"; Rao, K. R., Ed.; Plenum Press: New York, 1977; pp 115–130.
Chu, J. P.; Kirsch, E. J. *Appl. Microbiol.* **1972**, *23*, 1033.
Haque, A.; Scheunert, I.; Korte, F. *Chemosphere* **1978**, *7*, 65.
Honeycutt, R.; Adler, J. L. *J. Agric. Food Chem.* **1975**, *23*, 1097.
Ide, A.; Niki, Y.; Sakamoto, F.; Watanabe, J.; Watanabe, H. *Agric. Biol. Chem.* **1972**, *36*, 1937.
Kick, H.; Grosse-Brauckmann, E. *Z. Pflanzenernaehr., Dueng., Bodenkd.* **1961**, *95*, 52.
Kobayashi, K. In "Pentachlorophenol"; Rao, K. R., Ed.; Plenum Press: New York, 1977; pp 89–105.
Kuwatsuka, S.; Igarashi, M. *Soil Sci. Plant Nutr. (Tokyo)* **1975**, *21*, 405.
Lu, P.-Y.; Metcalf, R.; Cole, L. K. In "Pentachlorophenol"; Rao, K. R., Ed.; Plenum Press: New York, 1977; pp 53–63.
Munakata, K.; Kuwahara, M. *Residue Rev.* **1969**, *25*, 13.
Murthy, N. B. K.; Kaufmann, D. D.; Fries, F. *J. Environ. Sci. Health, Part B* **1979**, *B14*, 1.
Rott, B.; Nitz, S.; Korte, F. *J. Agric. Food Chem.* **1979**, *27*, 306.
Sandrock, K.; Attar, A.; Bieniek, D.; Klein, W.; Korte, F. *J. Labelled Compd. Radiopharm.* **1978**, *9*, 197.
Suzuki, T.; Nose, K. *Noyaku Seisan Gijutsu* **1971**, *2b*, 21.
Tashiro, S.; Sasmoto, T.; Aikawa, T.; Tokunaga, S.; Taniguchi, E.; Morifusa, E. *Nippon Nogei Kagaku Kaishi* **1970**, *44*, 124.
Weiss, U.; Scheunert, I.; Klein, W.; Korte, F. *J. Agric. Food Chem.* **1982**, following paper in this issue.
Wong, A. S.; Crosby, D. G. In "Pentachlorophenol"; Rao, K. R., Ed.; Plenum Press: New York, 1977; pp 19–25.

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