Distribution of ^{14}C and Arsenic Derived from [^{14}C]Cacodylic Acid in an Aquatic Ecosystem

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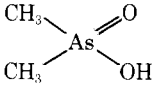
The distribution, accumulation, and degradation of the arsenical herbicide [^{14}C]hydroxydimethylarsine oxide ([^{14}C]cacodylic acid) was investigated in aquatic ecosystems containing three soil types, water, algae (*Oedogonium cardiacum*), duckweed (*Lemna minor L.*), snails (*Physa*, sp.), daphnia (*Daphnia magna*), catfish (*Ictalurus punctatus*), and crayfish (*Procambarus clarki*). After cacodylic acid incorporation, soils were flooded with 80 l. of water and allowed to incubate for 7 days, and ^{14}C and arsenic contents were determined intermittently in each component over the 60-day experimental period. ^{14}C from cacodylic acid increased in the water phase during the first 30–50 days of the experiment, reached a plateau, and finally declined. Accumulation of ^{14}C in aquatic organisms was quantitatively larger than arsenic accumulation, thus indicating degradation of cacodylic acid to various arsenical compounds in the aquatic ecosystem. Crayfish and snails contained higher arsenic and lower ^{14}C residues than algae, duckweed, and snails. Approximately 5, 31, and 48% of the arsenic initially added to the three soils was lost during the experiment. Bioconcentration ratios suggest that cacodylic acid does not accumulate greatly in the aquatic environment.

The organoarsenical herbicides constitute one of the last major metal or metalloid-containing pesticides being used commercially in the United States. Hydroxydimethylarsine oxide (cacodylic acid) is used as a contact herbicide in cotton producing areas and may supply arsenic to the aquatic environment through direct contamination, leaching, or run-off from treated fields. There is sufficient environmental concern over the alkylation and bioconcentration of metals and metalloids to warrant a detailed investigation of the distribution of arsenic and organoarsenicals in the aquatic environment.



Aquatic organisms may concentrate more arsenic than do terrestrial organisms. Reay (1972) reported concentration factors (based on concentration in plants/concentrations in water) of 100 to 20,000, on a dry weight basis, for various species of aquatic plants growing in the Waikato River in New Zealand. Arsenic concentrations in water varied from <10 μg of As/l. to about 75 μg of As/l. at the various sampling sites. However, high concentrations of arsenic (288 ppm of As) in aquatic plants were not toxic when fed to sheep (Lancaster *et al.*, 1971). Marine algae are capable of complexing arsenic (Chapman, 1926) and are probably the main route of entry of arsenic in fish (Lunde, 1972). Little inorganic arsenic is ingested directly from water, but is taken in with food and metabolized to both water-soluble and lipid-soluble organoarsenical compounds. Some inorganic arsenic present in water is absorbed directly by fish, but neither water-soluble nor lipid-soluble organoarsenical compounds were detected.

Various aquatic organisms concentrate or accumulate different amounts of arsenic. The amount of arsenic accumulated by fish varies from <1 ppm for most species to a high of 10 ppm, while crustacea may accumulate as much as 100 ppm of As (Chapman, 1926; Windom *et al.*, 1973). Fresh water crayfish (*Astacus pallipes*) generally accumulate less arsenic (1 ppm of As) while saltwater crayfish (*Palinurus vulgaris*) contained 30–40 ppm of As.

The persistence and reactions of [^{14}C]cacodylic acid in soils were studied by Woolson and Kearney (1973). Degradation of cacodylic acid in soils proceeds by two mechanisms. Anaerobically, a major portion was converted to a volatile organoarsenical and was lost from the soil system. Aerobically, about 35% was converted to a volatile organoarsenical compound while 41% was metabolized to $^{14}CO_2$ and arsenate within 24 weeks after application. In an aquatic system, either metabolic fate may occur.

In a previous study, the distribution of alkylarsenicals was measured in a model microecosystem (Isensee *et al.*, 1973). Lower food chain organisms (algae and *Daphnia magna*) accumulated more cacodylic acid and dimethylarsine (measured as ^{14}C) from water than did higher food chain organisms (snails and fish). The amounts of accumulation indicated that cacodylic acid did not show a high potential for biomagnification in the aquatic environment.

Due to the current concern over the distribution of pesticides in the environment, the accumulation of cacodylic acid in microecosystems was reexamined using different aquatic organisms. In the present experiments, the model ecosystem has been considerably reorganized to include bottom feeding organisms (catfish and crayfish) indigenous to cotton producing areas where most of the organoarsenical herbicides are being used.

METHODS AND MATERIALS

Synthesis of [^{14}C]Cacodylic Acid. ^{14}C-Labeled cacodylic acid was prepared by reacting [^{14}C]methyl bromide (0.3 m*M*; sp act., 14.7 μCi/mg) with methyldichloroarsine (1.55 m*M*) in sodium hydroxide (1.0 m*M*, 10 *N*) under nitrogen. The reaction was carried out at 85° in a sealed ampoule for 48 hr. The reaction mixture was applied to a silica gel column and [^{14}C]cacodylic acid was eluted using methanol-10^{-3} *M* ammonium hydroxide (8:2, v/v), 64% yield; sp act. 1.2 μCi/mg. Its identity was verified through cochromatography on tlc plates with authentic cacodylic acid.

Soil Treatment and Ecosystem Preparation. Three soils, Willacy sandy loam, Hidalgo clay loam, and Laredo silt loam, from a cotton producing region in Texas, were used (Table I). Duplicate 500-g (air dried) samples were treated with 15 ml of an aqueous solution containing 19 μCi of [^{14}C]cacodylic acid and 244 mg of unlabeled caco-

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Table I. Properties of Experimental Soils

Soil	pH	Clay, %	Silt, %	Sand, %	O.M., %	As, ppm
Hidalgo clay loam	7.7	41.2	15.2	43.6	2.3	9.5
Laredo silt loam	7.7	20.4	54.8	24.8	2.2	3.0
Willacy sandy loam	7.8	3.7	24.5	71.7	1.2	2.4

Table II. Analysis of Soil Treated with [^{14}C]Cacodylic Acid in an Aquatic Microecosystem

	ppm at day 0			ppm at day 59			% remaining	
	^{14}C[a]	(As)[a]	As[b]	^{14}C	(As)	As	^{14}C[c]	As
Hidalgo cl	21.2	(11.6)	10.6	2.4	(1.3)	3.1	11.8	29.2
Laredo sil	20.9	(11.4)	8.0	2.9	(1.6)	3.3	15.2	41.2
Willacy sl	20.6	(11.2)	9.6	2.8	(1.5)	4.6	13.6	49.5
						Av	13.5	40.0

[a] Concentration of cacodylic acid or (arsenic) based on ^{14}C analysis. [b] Soil As determined colorimetrically minus control soil As. [c] May contain metabolized ^{14}C.

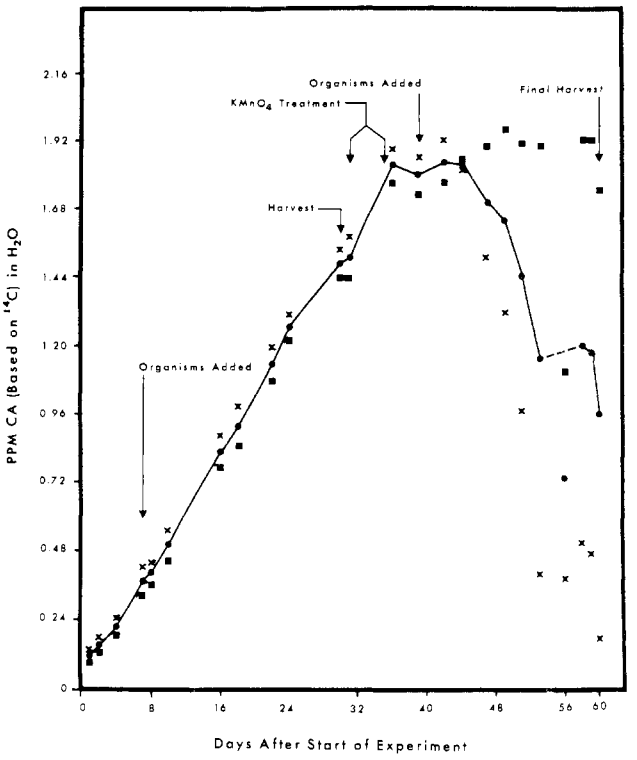

Figure 1. Cacodylic acid (based on ^{14}C measurement) in water from [^{14}C]cacodylic acid adsorbed to Hidalgo clay loam: (×) tank A; (■) tank B; (●) mean.

dylic acid (sp act. 0.076 μCi/mg). After the solution evaporated, the soil was mixed in a V-blender for 30 min, combined with 10.9 kg of the same soil, and mixed for an additional 30 min by rolling in a 20-l. carboy. After mixing, the soils (11.4 kg of each containing 21.4 ppm of cacodylic acid which approximately equals the concentration in surface 3.8-cm soil after application of highest rate used in the field) were layered on the bottom of separate 110-l. all-glass aquarium tanks, covered with pea-gravel and aluminum window screen, flooded with 80 l. of distilled water, and allowed to equilibrate for 1 week without aeration. Screen was also used to vertically bisect the tanks to protect the catfish from the predaceous crayfish. One control tank each of the Hidalgo and Laredo soils, containing all components minus cacodylic acid, was also prepared. No control for the Willacy soil was run since organisms survived during previous experiments (unpublished data). After 1 week, aeration was started and seven catfish (*Ictalurus punctatus*), three crayfish (*Procambarus clarki*), several hundred daphnids (*Daphnia magna*), ten snails (*Physa* sp.), several hundred milligrams of filamentous algae (*Oedogonium cardiacum*), and 10–20 duckweed (*Lemna minor L.*) plants were added to each tank. Water lost by evaporation was replaced as necessary.

Experimental Operation and Sampling. Two 1-ml water samples were taken from each tank at about 2-day intervals and radioactivity analyzed by liquid scintillation methods described in the subsequent sections. Additional 100-ml water samples, taken 8, 22, 38, 50, and 59 days after the soils were flooded with water, were analyzed for arsenite, arsenate, and total arsenic by colorimetric and atomic absorption spectrometry (AA) methods (Woolson *et al.*, 1971; Kan, 1973). Catfish and crayfish were fed brine shrimp and chopped perch, respectively, every 3–4 days.

Twenty-two days after the organisms were added, an infestation of parasites (*Lerneus* sp. copepods) was noted in all treated and control tanks and was considered to be responsible for an increasing crayfish mortality rate. The next day, all organisms were harvested and analyzed. One gram of $KMnO_4$ was added to each tank (about 12.5 ppm) 1 and 5 days after harvest to kill the parasites. New organisms were added (two crayfish, five catfish, five snails, about 200 daphnids, and several hundred milligrams of algae) 9 days after the first harvest. This second group of organisms was harvested after 20 days exposure. No parasites were observed during this second exposure period.

Sample Preparation and Analysis. Algae, duckweed, and daphnids were dried at 60° for 12 hr. Dry weights were taken, and samples were combusted at 1000° for 7 min in a stream of oxygen. Carbon dioxide from combustion was trapped in 10 ml of 2-methoxyethanol–ethanolamine (7:1). Five milliliters of this solution was mixed with 10 ml of scintillation cocktail (5 g of PPO + 150 mg of POPOP, + 1 l. of toluene) and counted for 10 min. Snails were homogenized in 10 ml of methanol in a glass, hand-held tissue grinder. Fingerling catfish were cut up before being homogenized with sufficient distilled water to make the homogenate approximately 0.1 g of tissue/ml. The soft tissue of the crayfish was dissected from the exoskeleton and homogenized as described for the catfish. Chiten from the carapace of the crayfish was digested with HNO_3–$HClO_4$–H_2SO_4 (20:1:4) for 4 hr and analyzed for total arsenic by atomic absorption spectroscopy. Aqueous homogenates were analyzed for ^{14}C by liquid scintillation counting (15 ml of scintillation cocktail containing 200 ml of 2-ethoxyethanol, 12 g of PPO, 600 mg of POPOP, and 60 g of naphthalene per liter of dioxane). Soils were dried at 100° overnight and mixed, and 1-g samples were weighed into 100-ml standard taper kjeldahl flasks. Both homogenates and soils were then analyzed for total arsenic colorimetrically as described by Woolson *et al.* (1971) or by AA. The AA analysis was performed after reduction to arsine, by adding $NaBH_4$ to a solution of *ca.* 20% HCl. The arsine gas was collected in a balloon along with the generated hydrogen and swept with argon into an argon–hydrogen flame of a Perkin-Elmer Model 303 AA spectrometer. Arsenic could be analyzed in the range of 0.05–1.0 μg of As. Samples of the soils were also combusted and analyzed, as described above, for total ^{14}C.

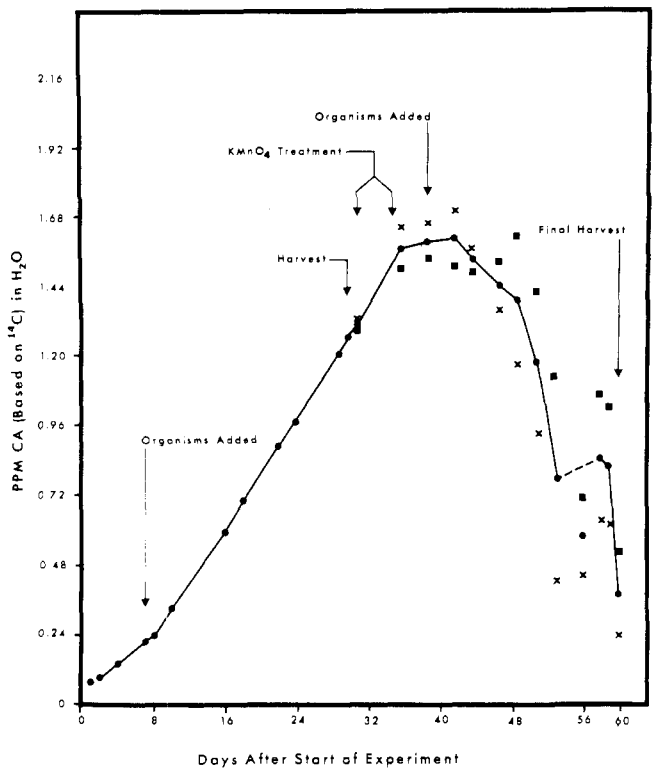

Figure 2. Cacodylic acid (based on ^{14}C measurement) in water from [^{14}C]cacodylic acid adsorbed to Laredo silt loam: (×) tank A; (■) tank B; (●) mean.

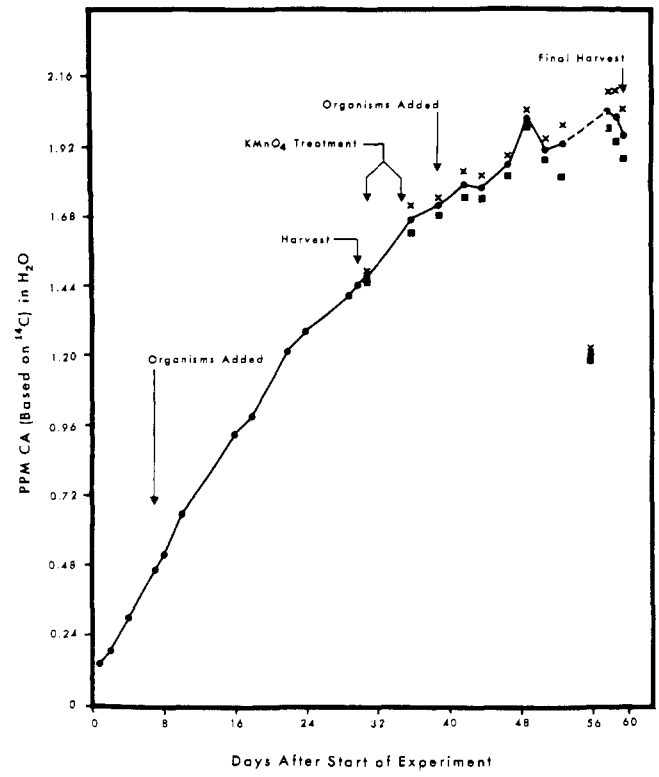

Figure 3. Concentration of cacodylic acid (based on ^{14}C measurement) in water from [^{14}C]cacodylic acid adsorbed to Willacy sandy loam: (×) tank A; (■) tank B; (●) mean.

Water from the tanks was analyzed directly for ^{14}C by using the dioxane cocktail and for total arsenic by using colorimetric or AA methods. The oxidative state of arsenic in water was determined by extraction of arsenite with benzene from acidified water (8 *N* HCl). Arsenate remained in the original aqueous layer. The arsenite was back-extracted with a phosphate buffer (pH 9). The arsenic content of each phase was determined colorimetrically. One liter of water from each tank was taken on the day of the second harvest, evaporated to 5 ml under vacuum at 40°, and examined by tlc using the methanol–ammonia solvent system. Autoradiographs were made by a 3-week exposure of the plates to No-Screen X-ray film.

RESULTS AND DISCUSSION

Soil was analyzed for ^{14}C after mixing and at the conclusion of the experiment (Table II). The per cent remaining was calculated relative to the initial amount found as ^{14}C or as total As after subtraction of As in the untreated soil. After 59 days, 13.5% of the ^{14}C radioactivity and 40.0% of the As from the cacodylic acid originally applied to soil remained (average of the three soils). These differential losses of As and ^{14}C indicate that, in soil, the C–As bond of cacodylic acid is being split as previously reported by Woolson and Kearney (1973).

Cacodylic acid concentration in water based on total radioactivity is presented in Figures 1, 2, and 3. The cacodylic acid in solution increased almost linearly throughout the first experiment (30 days). After $KMnO_4$ treatment, ^{14}C continued to increase, then leveled off, and finally decreased. In a separate experiment, $KMnO_4$ did not destroy cacodylic acid even when being heated during distillation of arsine. ^{14}C in solution for both replications of Laredo and one of Hidalgo decreased rapidly after 42–46 days while the second replication of Hidalgo and both replications of Willacy did not decrease until the very end of the experiment. The reasons for these differences between soils and replications in ^{14}C solution levels are unknown. However, different time periods for the buildup of microbial populations capable of degrading cacodylic acid may partly account for these erratic ^{14}C levels. Four of the six replications underwent a drastic reduction of ^{14}C in water, then recovered between 53 and 58 days. The authors have connected day 53 to 58 (bypassing day 56) with a dashed line to signify that we have no explanation for this change even though we are confident that the ^{14}C measurements are correct (Figures 1, 2, and 3). The reduction in ^{14}C levels may be indicative of cacodylic acid degradation to $^{14}CO_2$ and arsenate or reduction to methylarsines and loss through volatilization. The $^{14}CO_2$, if produced, would be available for uptake by the organisms. Tlc analysis of the water after 59 days failed to show the presence of cacodylic acid. The ^{14}C in the water was associated with volatile chemical forms, possibly $^{14}CO_2$ or organoarsenical compounds, which were lost during evaporation since no activity was found in water after concentration.

The chemical distribution of arsenical forms in water is presented in Table III. The ^{14}C increased to a maximum between days 38 and 50 and then declined. The total As in water from cacodylic acid treatment (difference between treated tanks and control tanks) generally increased for 2–3 weeks and then leveled off. Very little arsenite was present at this time. Amounts of arsenite were nearly identical in both treated and control tanks and remained essentially constant after 22 days. Water concentrations of ^{14}C from the Laredo and Hidalgo soil tanks were not different. The ^{14}C level in both reached a maximum, and then declined. The concentration of ^{14}C in water from the Willacy soil increased throughout the 59 days. Water concentrations of arsenate were essentially the same for the Laredo and Hidalgo soils, but lower for the Willacy soil. Arsenate concentration increased significantly by day 50. These observations, along with a decrease in ^{14}C in solution, add strength to the conclusion that the cacodylic acid was degraded in at least two of the three soils.

Problems were encountered in keeping aquatic organisms alive until harvest. An infestation of parasites accounted for some crayfish mortality in both treated and untreated tanks. No differences in survival of catfish or crayfish between treated and control tanks were observed

Table III. Distribution of Arsenite, Arsenate, Total Arsenic, and Cacodylic Acid in Water at Five Sampling Dates

		Water concn, ppm								
		Hidalgo			Laredo			Willacy		
Day	Species	+CA	−CA[a]	Δ[b]	+CA	−CA	Δ	+CA	−CA	Δ
8	Arsenite	0.14	0.02	0.12	0.03	0.02	0.01	0.05	ND	ND
	Arsenate	0.07	0.07	0.00	0.05	0.03	0.02	0.06	ND	ND
	Total As[c]	0.38	0.23	0.15	0.20	0.07	0.13	0.19	ND	ND
	[^{14}C]CA[d]	0.22	0	0.22	0.13	0	0.13	0.29	ND	0.29
22	Arsenite	0.13	0.08	0.05	0.10	0.08	0.02	0.09	ND	ND
	Arsenate	0.15	0.09	0.06	0.09	0.08	0.01	0.10	ND	ND
	Total As	0.73	0.12	0.61	0.54	0	0.54	0.63	ND	ND
	[^{14}C]CA	0.61	0	0.61	0.47	0	0.47	0.65	ND	0.65
38	Total As	0.64	0.12	0.52	0.36	ND[e]	ND	0.40	ND	ND
	[^{14}C]CA	0.96	0	0.96	0.86	0	0.86	0.92	ND	0.92
50	Arsenite	0.11	ND	ND	0.14	0.14	0.00	0.13	ND	ND
	Arsenate	0.55	ND	ND	0.54	0.12	0.42	0.19	ND	ND
	Total As	0.80	0.12	0.68	0.51	0.12	0.39	0.66	ND	ND
	[^{14}C]CA	0.77	0	0.77	0.63	0	0.63	1.02	ND	1.02
59	Arsenite	0.08	0.08	0.00	0.12	0.12	0.00	0.08	ND	ND
	Arsenate	0.50	0.15	0.35	0.52	0.13	0.39	0.08	ND	ND
	Total As	0.82	0.12	0.70	0.51	0.13	0.38	1.00	ND	ND
	[^{14}C]CA	0.52	0	0.52	0.20	0	0.20	1.05	ND	1.05

[a] Soil minus cacodylic acid, or the control soil. [b] Difference between treated and control soil; therefore represents the arsenic contribution to water from cacodylic acid treatment. [c] Total As by digestion with HNO_3-H_2SO_4-$HClO_4$ digestion (Woolson *et al.*, 1971). [d] Cacodylic acid as determined by ^{14}C analysis, but expressed as arsenic. At later dates, this value may not reflect cacodylic acid. due to metabolism to $^{14}CO_2$. [e] ND, not determined.

Table IV. Concentration of [^{14}C]Cacodylic Acid and Arsenic in Tissue (ppm) from Aquatic Organisms in a Microecosystem

	Organisms analyzed						
						Crayfish	
Soil	Algae	Duckweed	Daphnids	Snails	Catfish	Soft tissue	Chitin
	Parent Compound (Based on ^{14}C for First Harvest)						
Hidalgo	119.1	77.5	75.7	39.1	1.13	0.8[b]	ND
BR[a]	80	52	51	26	0.8	0.5	ND
Laredo	387.0	114.6	215.3	32.1	3.65	2.6[c]	ND
BR	298	88	147	25	2.8	2.0	ND
Willacy	148.4	85.2	ND[e]	32.5	1.06	2.9[d]	ND
BR	101	58	ND	22	0.7	2.0	ND
	Arsenic						
Hidalgo	ND	ND	ND	18.0[f]	0.0	3.3	1.1
BR	ND	ND	ND	23.0[g]	0.0	4.2	1.4
Laredo	ND	ND	ND	0.0	0.0	2.8	0.4
BR	ND	ND	ND	0.0	0.0	5.2	0.7
Willacy	ND	ND	ND	3.8	0.2	3.1	3.5
BR	ND	ND	ND	7	0.4	4.7	5.3

[a] Concentration of ^{14}C in tissues/concentration of ^{14}C in water = BR. [b] Two dead at harvest in control tank. [c] Two dead at harvest in control tank; two of three in A tank. [d] Two dead at harvest in A tank. [e] Not determined. [f] Arsenic in organisms from control tank subtracted. [g] Concentration of As in tissue/concentration of As in water.

in initial harvest. All organisms were harvested alive at the second harvest. A few dead crayfish were included in the analysis of the first harvest.

Concentrations of ^{14}C and arsenic in various biota and bioaccumulation ratios (BR) for the first harvest of organisms are presented in Table IV. Bioaccumulation ratios are calculated by dividing the ^{14}C or arsenic contents in the tissue by the ^{14}C or arsenic present in water at the time of harvest. Bioaccumulation ratios decreased as the position of the organism in the food chain increased. Thus, algae and duckweed have the highest amount of ^{14}C, whereas catfish and crayfish have very low levels. Bioaccumulation ratios for total arsenic are lower than those determined by ^{14}C analysis except for crayfish. The high ^{14}C residues in snail could be expected to reflect ^{14}C levels found in algae since snails are direct consumers of

Table V. Concentration of [^{14}C]Cacodylic Acid and Arsenic in Tissue (ppm) from Aquatic Organisms in a Microecosystem

	Organisms analyzed						
						Crayfish	
Soil	Algae	Duckweed	Daphnids	Snails	Catfish	Soft tissue	Chitin
	Parent Compound (Based on ^{14}C for Second Harvest)						
Hidalgo	2,378	1191	ND[a]	ND	8.1	5.7	ND
BR	2,483[b]	1244	ND	ND	8.5	5.9	ND
Laredo	10,123	1301	ND	394	103[c]	51.0	ND
BR	26,959	3466	ND	1050	275	13.6	ND
Willacy	318	174	ND	8.4	4.1	5.8	ND
BR	163	89	ND	4.3	2.1	3.0	ND
	Arsenic						
Hidalgo	12[d]	ND	ND	ND	0	0	0.1
BR	17[e]	ND	ND	ND	0	0	0.1
Laredo	2	0	ND	8	0	17.1	2.7
BR	5	0	ND	21	0	16.0	2.5
Willacy	3	3	ND	2	0	0	0.9
BR	3	3	ND	2	0	0	0.9

[a] ND, not determined. [b] Concentration of ^{14}C in tissue/concentration of ^{14}C in water. [c] Dead at harvest. [d] Background As subtracted. [e] Concentration of As in tissue/concentration of As in water.

Table VI. Balance Sheet of Arsenic from [^{14}C]Cacodylic Acid Treated Soil in a Microecosystem

Soil	Total As added, mg	mg of As remaining in[a] H_2O	mg of As remaining in[a] Soil	As lost, mg	As lost, %
Hidalgo cl	132	56	35	41	31
Laredo sil	130	30	38	62	48
Willacy sl	128	70	52	6	5

[a] Arsenic in water and soil from control tanks subtracted.

algae. Considering the number of organisms analyzed and natural variability, differences between BR's measured by ^{14}C or arsenic for catfish and crayfish are not significant for the first harvest.

Concentrations of ^{14}C and total arsenic and BR values for the second harvest of organisms are presented in Table V. These ^{14}C bioaccumulation ratios were found to be much higher than in the first harvest organisms. The lack of correlation between the high ^{14}C ratios and the much lower arsenic values does not support bioconcentration of cacodylic acid, although this could be occurring to a very limited extent. Rather, the ^{14}C data lend support to cacodylic acid degradation with subsequent uptake of ^{14}C by the plant life. Differences between BR's measured by the two methods are much larger for the second harvest. Autoradiography of a tlc of snail and algae extracts failed to show any parent cacodylic acid stored in the organisms.

A balance sheet for As in the model ecosystem is presented in Table VI. Arsenic values in check soils and water have been subtracted to give values that are the result of cacodylic acid treatment only. Significant amounts of arsenic were lost from the Hidalgo and Laredo soil systems. Little arsenic was lost from the Willacy soil tank. A garlic-like odor was observed above these tanks on day 25. The loss of arsenic from the total system was observed previously for cacodylic acid treated soils (Woolson and Kearney, 1973), and may be as dimethylarsine since it has a garlic-like odor. Quantitative differences in arsenic lost may reflect different populations, in each soil, of microorganisms responsible for the reduction of cacodylic acid to a volatile organoarsenical compound or due to differences in the ability of the soils to retain arsenical compounds. Arsenic in the biomass was an insignificant sink in relation to the total arsenic in the system.

In summary, lack of high levels of arsenic in catfish and crayfish indicates that cacodylic acid does not bioaccumulate in these members of the aquatic environment. Degradation of cacodylic acid to ^{14}C-containing products and inorganic arsenate does occur, while some reduction to volatile organoarsenical compounds may account for loss of arsenic from the system.

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