### Effects of Herbicide on the Environmental Ecosystem in Subtropics

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#### **ABSTRACT**

Herbicides play a very important role in modern agriculture. However, the herbicide applied to the agricultural field may accumulate in the field, converting the advantages to environment pollution. Many small animals in the ecosystem such as alderfly, earthworm, butterfly, loach, frog, firefly, some birds and aquatic organisms have been known to disappear gradually. In addition, several behavior of herbicides including adsorption by soil, movement by water, photodecomposition, volatilization to air, absorption by plant, metabolism by soil microorganisms and so on, are proceeded while the herbicide remained in the environment. In this review, fate and behavior of herbicides in the environment and their effect on ecosystem after their application are focused on four aspects: the first is the absorption and metabolism of herbicides by plant; the second is the residues of herbicides in soil and water environments; the third is the accumulation and release of herbicides in aquatic organisms and the fourth is the translocation of herbicides in model agricultural ecosystem. Many factors may affect the behavior and fate of herbicides after their application, climatic conditions and soil properties seem to be the most important. Therefore, the fate and behavior of herbicide in Taiwan, located on subtropical region, may differ from those in Korea.

Key words: Herbicide, Environment, Fate and Behavior, Ecosystem

Relationship between herbicides and ecosystem environment is found as follows. Environmental pollutants such as herbicides may be absorbed into plants or taken in by aquatic organism. The amounts of the pollutant transfer into organism are affected by the concentration of the pollutant residue in the environment. The organisms which survive in a certain level of pollutant concentration in environment will accumulate the pollutant to a maximum concentration within a shortterm of several days. Concentrations of the pollutant

residue in the organisms still remaine in the same level until the pollutant concentrations in environment are changed.

Pollutants residue in animals disappear more easily than that in plants, because metabolism only works in plants, but for animals both metabolism and excretion works. Aquatic organisms accumulate herbicide to a maximum concentration within 3 to 5 days, but after moving to clean water, the concentration of herbicide in organisms rapidly decreases to a negligible level within 3 to 5 days.

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Microorganisms play important roles in the dissipation of herbicide both in soil and water environments. The herbicide decomposes to produce non-toxic CO<sub>2</sub> for photosynthesis use, produces the metabolites with unknown toxicity. The rate of accumulation and clearance of herbicide by organism depends upon the fat contents. Due to the bio-magnification by food chain, the pollutant with higher lipid solubility gives the greater bio-concentration factor(BCF).

The present review primarily reports our recent studies on the behavior of herbicides in the subtropical environment after their use.

# ABSORPTION AND METABOLISM OF HERBICIDES BY PLANTS

According to the study on the effects of para-

quat (1,1'-dimethyl-4,4'-bipyridinium ion) on the growth of rice and corn seedlings, paraquat had no significant effect on the germination of rice and corn in a soil culture, but it was delayed for several days in a water culture. The growth of both stem and root in rice was inhibited by paraquat, and the growth of stem, but root in corn was not inhibited<sup>1)</sup>. Herbicide isouron [3-(5-tertbutyl-3-isoxazolyl)-1,1-dimethylureal was absorbed by the rice roots and translocated rapidly to the shoot(Table 1). Isouron was converted to eight known metabolites in the rice seedling. The critical concentration of isouron that inhibited the growth of rice seedling was about 0.10 ppm<sup>2</sup>. Uptake of isouron by sugarcane plants from soil was more rapid during the first 7 weeks than during the later stage. Isotope studies showed more than 83% of the radioactivity translocated to leaves

**Table 1.** Distribution of radioactivity in rice seedlings after incubation in 0.5 ppm of <sup>14</sup>C-isouron solution for various periods<sup>2)</sup>.

T 1 41 .			Radioac	tivity				
Incubation period	Whole	plant	Shoo	ots	Roo	Roots		
(h)	Total act. (dpm)	Specific (dpm/mg)	(dpm/mg)	(%)	(dpm/mg)	(%)		
1	8012	164	300	91.2	29	8.8		
6	8828	276	570	82.6	80	17.4		
24	11197	366	657	72.7	167	27.3		
72	12600	403	721	65.8	218	34.2		
168	20579	472	679	60.4	323	39.6		

Table 2. Distribution of radioactivity in the individual sugarcane plant<sup>3)</sup>.

<b>.</b>	Specific activity, Cpm/g.dry Wt (%)									
Treatment	Leaf	Scale	Stem	Root	Whole plant					
1 day	826(31.7)	1,097(33.2)	1,060(8.7)	870(26.4)	926(100)					
3 day	4,946(50.1)	2,744(12.8)	4,140(5.4)	4,207(31.7)	4,231(100)					
1 WK	9,571(63.5)	6,596(18.8)	5,838(4.9)	6,384(12.8)	8,112(100)					
2 WKS	20,619(83.5)	9,267(7.3)	7,157(1.9)	11,382( 7.3)	17,418(100)					
4 WKS	48,675(84.8)	19,218( 5.3)	10,728(1.9)	10,132( 7.9)	33,839(100)					
7 WKS	54,645(86.4)	25,469( 7.7)	12,093(2.4)	9,575( 3.5)	41,273(100)					
16 WKS	5,745*(87.2)	-	1,723(3.4)	2,887( 9.4)	4,751(100)					
25 WKS	2,425**(87.6)	-	574(8.5)	3,143( 3.9)	1,894(100)					

<sup>\*</sup> Dead leaf 16,528 cpm/g. dry wt.(34.4%).

<sup>\*\*</sup> Dead leaf 7,585 cpm/g. dry wt.(48.2%).

after 2 weeks. Larger amount of isouron was present in the young leaves at the earlier stage, but most of the activity was found in the old leaves at the late stage after 7 weeks(Table 2)<sup>3)</sup>.

The selectivity of butachlor [2-chloro-2',-6'diethyl-N-(butoxymethyl)-acetanilide] on rice and barnyardgrass was found in the early stage of germination, the selectivity was not very distinctive between the two species when treated at 5-leaf stage of development<sup>4)</sup>. Glyphosate [N-(phosphono methyl) glycin] did not affect the germination of rice seeds up to a concentration of 50 ppm. No difference was observed between Tainan No. 5 (Japonica-type) and Taichung No. 3(Indica-type). But the growth of rice seedling in nutrient solution was affected at concentrations of 1.0 and 2.5 ppm for Tainan No. 5 and Taichung No. 3, respectively<sup>5)</sup>. Naproanilide [2-(2-naphthyloxy) propionanilide] is translocated easily to the upper part of tobacco plants(Table 3), when compared to that of rice, the translocation rate in tobacco is much higher and may therefore give a possible mechanism of herbicidal selectivity. At least seven metabolites

have been identified in tobacco callus<sup>6)</sup>.

## RESIDUES OF HERBICIDES IN SOIL AND WATER ENVIRONMENTS

Small variations of dissipation of radioactivity for <sup>14</sup>C-2,4,-d [2,4-dichlorophenoxy) acetic acid] in higher and lower concentrations and in different samples of river water (Hsin-Tien river, Ta-Tu river, Err-Jin river and Li-U river located in the northern, central, southern and eastern parts of Taiwan, respectively) were found. But about half of glyphosate disappeared from water samples of original concentration at 100ppm and only 11 to 22% remained at that of 100 ppb after 56 days incubation for all river water samples except the sample from Hsin-Tien river. More than 80% of paraquat remained in water samples<sup>7</sup>.(Table 4)

Degradation of isouron in soil followed the first kinetic with half-lives of 42 to 203 days under various conditions in the laboratory experiments (Table 5). More than six degradation products were found in the soil<sup>8</sup>.

Table 3. Distribution of radioactivity derived from <sup>14</sup>C-naproanilide in tobacco plants<sup>6</sup>.

Days after	Ra	dioactivity, dpm/mg, dry W	t.*
treatment	Root	Stem	Leaf
7	4,843(0.88%)	1,282(0.24%)	911(0.03%)
14	5,531(1.71%)	5,857(1.86%)	3,453(2.32%)

<sup>\*</sup> Percentages are related to the total radioactivity applied.

**Table 4.** Dissipation of 2,4-D, glyphosate and paraquat in river water after 56 days Incubation<sup>7</sup>.

	Residues of radioactivity or herbicide(%)									
River	2,4	-D	Glyph	osate	Paraquat					
Rivei	100ppm	100ppb	100ppm	100ppb	100ppm					
N	76.0	75.2	45.3	45.9	81.3					
E	81.1	88.7	47.6	10.8	83.1					
S	80.0	84.2	54.4	21.6	84.9					
C	77.5	85.5	38.4	15.2	81.2					

N=Hsin-Tien river(North part); S=Err-Jin river(South part); E=Li-U river(East part); C=Ta-Tu river(Central part).

**Table 5.** Half-lives of isouron under different conditions in laboratory experiments at an application rate of 4 ppm<sup>8)</sup>.

Temp.	Moisture	Half-life (days)						
(°C)	(% of field capacity)	Touliow sandy loam	Taichung loam	Annei silty loam				
10	60	200	203	148				
20	60	84	79	67				
30	60	70	68	58				
40	60	61	56	42				
20	20	125	129	140				
20	40	97	92	90				
20	90	73	75	59				
30	60	200*	208*	148*				

<sup>\*</sup> Autoclaved.

Isouron(I) was mineralized to 40 and 31% in sewage samples at 0.01 and 0.1 ppm, respectively,

but only 7% were mineralized in river-water samples in a 120-day period(Fig. 1). Although most of the radioactivity remained, only 66 to 68 % of the initial isouron were found in the riverwater samples. Major metabolites were found to be  $3-(5-tert-butyl-3-isoxazolyl)-1-mthylurea( <math>\coprod$  ) and 3-amino-5-tert-butyl-isoxazole(III) in the sewage samples<sup>9)</sup>. Isouron was relatively easily leached from the soils in the laboratory experiments using three typical sugarcane soils in Taiwan. Leaching with 398 mm of water at a rate of 8 mm/hr in the soil columns removed 87.7, 69.2 and 52.2% of surface-applied isouron from Touliow sandy loam, Taichung loam and Annei silty loam, respectively(Table 6). Most of the herbicide remained in the upper 30cm of the soil after receiving 347mm of rainfall for 112 days<sup>10)</sup>.

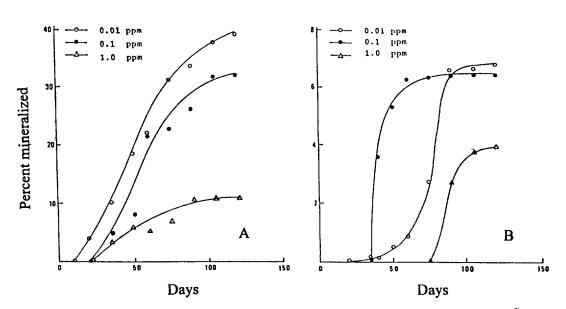


Fig. 1. Mineralization of isouron in sewage (A) and river water (B) at different concentrations<sup>9)</sup>

**Table 6.** Distribution of radioactivity in the soil columns leached with 398mm of water at a rate of 8mm/hr<sup>10</sup>.

Soil column	Ra	dioactivity(	<del>%</del> )
depth (cm)	Touliow sandy loam	Taichung loam	Annei silty loam
0-3	0.89	3.23	2.55
3-6	1.00	2.50	2.36
6-9	1.24	2.27	2.40
9-12	1.00	2.04	2.78
12-15	1.00	2.71	3.08
15-18	1.12	3.25	4.24
18-21	0.96	3.68	5.07
21-24	1.17	4.37	5.49
24-27	1.17	4.04	7.36
27-30	1.22	5.25	6.22
Total	10.77	33.34	41.55
Leachate	87.80	69.20	52.20
Total	98.57	102.54	93.75

When glyphosate irradiated under the UV-light of 254nm from a distance of 10cm, in a thin film as a solid state and in the aqueous solution for 72 hr, the major photodegradation product was AMPA[(aminomethyl) phosphonic acid]. The half life was about 8 hr, both in solid state and in aqueous solution. In the aqueous solution, 25.8% of glyphosate was decomposed in the dark after

72 hr<sup>11</sup>. More than 95% of naproanilide remained unchanged after irradiation for 72 hrs under UV lamp at dry state. In dark incubation, 8%(pH 6.5), 13%(pH 10) and 20%(pH 3) of naproanilide were degraded in the aqueous solution after 72 hrs. Photodegradation rate of naproanilide in the aqueous solution at 35°C was more rapid than that 25 and 15°C, and faster at a higher initial concentration than that at the low initial concentration. More than twelve photodecomposition products were detected in the solution after UV irradiation.

Simazine [2-chloro-4,6-bis(ethylamino)-s-triazine] had a half-life of 18 days in the summer which is relatively hot and wet, a half of 24 days in the cooler and dryer winter season of the Taichung area, But in the medium temperature and medium precipitation of the autumn season of the Taipei, area, the half-life was found to be 14 days. These results indicate that climate was not the only factor affecting degradation of simazine, but soil properties may also be related to the degradation rate <sup>13)</sup>. The soil properties and standard half-lives carried out in different countries from 16 location sites in the international collaborative experiments are shown in Table 7.

Table 7. Soil properties and half-lives of simazine at different location of sites<sup>13)</sup>.

Location of site	S		Half-life	
Location of site	Organic carbon (%)	Soil texture	pН	(day)
Saskatchewan, Canada	4.00	Clay	7.7	101
Uppsala, Sweden	3.60	Clay	6.5	88
Alberta, Canada	1.26	Clay loam	7.8	88
Ontario ( I ), Canada	0.52	Sand	5.2	63
Ontario (II), Canada	1.50	Loamy sand	5.6	63
Braunschweig, Germany	0.99	Loam	6.5	54
Wageningen, Holland	2.38	Sand	5.6	51
Warwik, England	1.30	Sandy clay loam	6.6	46
Firenze, Italy	0.98	Sandy loam	6.7	39
British Columbia, Canada	0.71	Loamy sand	7.5	38
Harpenden, England	1.75	Clay loam	7.5	37
Oxford, England	2.10	Sandy loam	5.8	31
Taichung, Taiwan(Winter)	0.83	Clay loam	5.2	24
Maarn, Hollan	1.40	Sand	5.6	21
Taichung, Taiwan(summer)	0.83	Clay loam	5.2	18
Tapei, Taipei	1.04	loam	4.3	14

Photodecomposition of butachlor in aqueous solution was more complicated. Under UV light the half-life was found to be about 0.8hr, but under sunlight it was 5.4 hr. At least 24 photodecomposed products were obtained from the aqueous solution(Fig. 2)<sup>14</sup>.

The effects of the herbicide butachlor on ammonification and nitrification were studied by adding 100 ppm NH<sub>2</sub>-N as CO(NH<sub>2</sub>)<sub>2</sub> or NH<sub>4</sub>-N as (NH<sub>4</sub>)<sup>3</sup>SO<sub>4</sub> at pH 6.8 by the addition of CaCO<sub>3</sub>

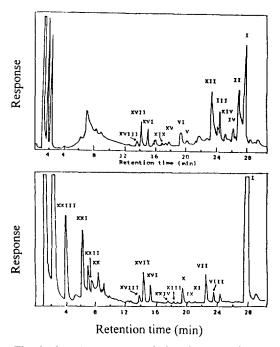


Fig. 2. Gas chromatogram of photodecomposed products from butachlor in aqueous solution under UV lamp (above) and sunlight (below) for 24 hr. 14)

to the soil. Butachlor at all doses inhibited the rate of ammonification and stimulated the rate of nitrification slightly at  $30^{\circ}$ C when pH was adjusted from the original 4.9 to  $6.8^{15}$ ).

A leaching experiment on a soil column showed that most of the nitrofen [2,4-dichloro-1-(4-nitrophenoxy) benzene] remained in the upper 3cm of the soil column after leaching with 0.05 M CaSO<sub>4</sub> solution for 56 days (100m/day). Nitrofen may not contaminate the ground water based on the results evaluated by the ground water pollution potential model (GWP model) (Table 8)<sup>18)</sup>. Evaluation by GWP model showed that 2,4-dichlorophenol will contaminate the groundwater at a depth of 10m in Pingchen soil after 16.4 or 8.2 years with infiltration rate of 50 or 100cm/year, respectively (Table 8)<sup>18)</sup>.

The residues of three herbicides butachlor, benthiocarb [S-(4-chlorobenzyl)-N, N-diethylthiocarbamate] and chlomethoxynil (2,4-dichlorophenyl-3'-methoxy-4'-nitro phenyl ether) in irrigation and drainage water of paddy field after application were investigated at northern (I-Lan prefecture), central (Tao-yuan prefecture) and southern (Yun-Lin prefecture) parts of Taiwan. Evaluation of the residues of butachlor and benthioncarb in paddy water on danger level to carp are also studied. The results showed that the residue of the herbicides were much lower than that of the safe concentration to carp both in field and drainage water 18.

**Table 8.** Evaluation of residual mass and travel time of herbicide nitrofen leaching to 3cm depth of ground water and several chlorophenol compounds to 10m depth by using GWP model<sup>16,17)</sup>.

	Average	Lukang sil	ty clay loam	Pingch	Pingchen clay		
	Drainage (m/year)	Travel time (year)	Residual mass (%)	Travel time (year)	Residual mass (%)		
Nitrofen	3	490	0	22.5	0		
Nitrofen	10	147	0	6.7	0		
2.4-Dichlorophenol	1	64.6	0	8.2	2.85		
2.4.6-Trichlorophenol	1	161	0	88.5	0		
Pentachlorophenol	1	235	0	175	0		

#### ACCUMULATION AND RELEASE OF HERBICIDE IN AQUATIC ORGANISMS

Accumulation and release of herbicide butachlor, thiobencarb and chlomethoxyfen by fish and mussel in the aquatic environments at lower concen-

trations were studied. Experiments were performed by exposing fish, clam and shrimp to the herbicide at a concentration of one hundredth and one thousandth of their respective 48-hr LC<sub>50</sub> values. Macrobranch shrimp showed higher tolerance than all other organisms. Maximum accumulation in

Table 9. Maximum accumulation (within 3 to 5-days exposure) in organisms and the bioconcentration factor(BCF)19)

		Butachlor			Thiobencarb	
Organism	A	В	BCF	A	В	BCF
Carp	18.0	195	10.8	37.2	2,160	58
	9.0	51	5.7	18.6	718	39
Tilapia	10.0	24	2.4	20.0	487	24
	1.0	7	7.0	2.0	21	10
Loach	10.0	28	2.8	20.0	613	31
	1.0	7	7.0	2.0	27	13.5
Grass carp	2.5	143	57	15.0	124	8.3
	1.25	133	106	7.5	90	12.0
Eel	5.0	255	51	10.0	159	16
	2.5	198	79	5.0	92	18
Black silver carp	2.4	235	98	5.0	1,876	375
	6.4	88	220	1.4	1,223	874
Freshwater clam	100.0	112	1.1	200.0	570	2.9
	16.0	60	6.0	20.0	145	7.3
Macrobranch	100.0	0.56	0.01	50.0	0.27	0.01
shrimp	10.0	0.30	0.03	5.0	0.03	0.01

A=Herbicide in water,  $\mu g/L$ .

B=Maximum accumulation in organism,  $\mu g/L$ .

Table 10. Excretion of herbicides butachlor and thiobencarb by organisms<sup>19)</sup>.

		Butachlór			Thiobencarb	
Organism	A <sup>1)</sup>	$\mathbf{B}^{2)}$	BCF <sup>3)</sup>	A <sup>1)</sup>	B <sup>2)</sup>	BCF <sup>3)</sup>
Com	10.0	44	14.0	20.0	1,263	446
Carp	1.0	6.7	3.5	2.0	24	9.9
T:1i-	10.0	25	9.6	20.0	432	37
Tilapia	1.0	6.6	3.8	2.0	20	4.1
Land	10.0	29	10.4	20.0	285	32
Loach	1.0	7.0	4.4	2.0	13.5	3.7
C	2.5	124	— <sup>4)</sup>	15.0	88	_
Grass carp	1.25	96		7.5	74	_
T-1	5.0	205	_	10.0	129	
Eel	2.5	171		5.0	92	_
Diagle gileran sam	2.4	475	334	5.0	3,687	1,900
Black silver carp	0.4	247	219	1.4	1,721	1,160
Frankricker alam	100.0	132	14.0	200.0	582	126
Freshwater clam	10.0	62	10.3	20.0	130	19
Macrobranch	100.0	0.1	_	50.0	_	_
shrimp	10.0	0.1		5.0		_

4)  $< 0.01 \mu g/L$ .

<sup>1)</sup> Herbicide concentration in water for 3 days of organisms prefeeding.
2) Herbicide concentration in organism after removing to clean water for 0 day.
3) Herbicide concentration in organism after removing to clean water for 5 days.

organisms and the BCF at long-term test are shown in Table 9. The results of depuration tests are shown in Table 10. More than half of the herbicide residue in carp, tilapia, loach, clam and black silver carp was excreted within 5 days in clean water<sup>19)</sup>.

Accumulation and relative concentration of 2,4-D and glyphosate in carp and tilapia were studied by using labelled and unlabelled chemicals. About 83 (at a concentration of 0.5 ppm) and 91% (of 0.05 ppm) of the radioactive matter remained in the water until 14 days after <sup>14</sup>C-2,4-D amended, but only 17.2% of glyphosate remained in the water with 0.05 ppm concentration of glyphosate. No significant variation was shown in the accumulation of herbicide in fish from 2 to 7 days. Although glyphosate disappeared within 3 days in water under sunlight, the radiochemicals in the water hyacinth remained constant up to the 14th day<sup>20)</sup>. Applying glyphosate to control water hyacinth in ponds and in irrigation canals was also studied. The water hyacinth in ponds and irrigation canals was controlled at the rates applied, and residue of the herbicide in the aquatic environment after the treatment was found to be less than 1 ppm. Moreover, glyphosate dissipated rapidly from the aquatic environment, with less than 22% of the maximum concentration remaining 1 day after the treatment in the ponds; no detectable residue was found in the canals 4 hr after treatment<sup>211</sup>.

# TRANSLOCATION OF HERBICIDE IN MODEL AGRICULTURAL ECOSYSTEM

A model ecosystem was first developed by Metcalf et. al. in 1971 to evaluate pesticide biodegradability and ecological magnification in a sorghum ecosystem. A modification by Hsu allowed substitution of biota from paddy field in Taiwan in 1979. Seven metabolites were identified and two unknown compounds were detected in the study on the metabolic fate of naproanilide by

Table	11	Dietribution	οf	radioactivity	in	the	components	of	the	model	ecosystem <sup>22</sup>	2)

Components	Fresh weight (g)	Total activity (dpm)	% of Application	Specific Act. (dpm/g)	Ecological magnif
Organisms			1.5		
Rice plant	6.86	16,523	0.258	2407	_
Planthopper	0.1958	145	0.003	741	and the same of th
Wolf spider	0.3165	500	0.008	1580	
Grasshopper	0.9918	923	0.014	932	
Alga	15.68	28,587	0.447	1822	36.5
Giant duckweed	8.39	11,224	0.175	1337	26.7
Water flea	0.2858	t <sup>a)</sup>	_		_
Mosquito larva	0.2178	158	0.003	725	_
Paddy snail	4.49	17,338	0.271	3858	96.5
Mosquito fish	2.64	19,433	0.304	7353	147.0 <sup>b)</sup>
Water (Total 10L)	500.0	25,000	7.8	50	1.0
Sand (Total 10kg)	500.0	250,000	78.1	500	_

a) t: trace (<50dpm)

b) EM values of chlorinated hydrocarbon insecticides for mosquito fish: p,p'-DDT 88,273; aldrin 4,495; dieldrin 1,900; and  $\gamma$ -BHC 1,475.

using a C<sup>14</sup>-labelled compound in rice paddy model ecosystem in the laboratory. In a 23-day experiment, 78.1, 7.8 and 1.5% of the total radio-activity were found in the sand, water and biota, respectively, for the herbicide naproanilide in a rice paddy model ecosystem (Table 11). The ecological magnification (EM) values were much smaller and the biodegradability indices (BI) values were much greater than those of persistent chlorinated hydrocarbon insecticides for boita<sup>22</sup>.

Microbial degradation by mineralization or cometabolism was determined by chemical concentration and environment. Herbicides diuron [3-(3,4dichlorophenyl)-1, 1-dimethylurea] and linuron [3-(3,4-dichlorophenyl)-1-methoxy-1-methylurea] were mineralized when added to sewage at a concentration of 0.5 ppb, but not at 2.0 ppm. No evidence for cometabolism of the higher levels of these two herbicides was obtained, but significant amount of an unknown product appeared at the lower diuron levels<sup>23)</sup>. Herbicide IPC(isopropyl N-Phenylcarbamate) at 400 pg/ml and 1 µg/ml was mineralized in samples of sewage, but only the lower concentration was mineralized in lake water samples in a 50-day period. IPC at 1 µg /ml disappeared from lake water, but it was converted to organic products (Table 12)24).

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Table 12. Distribution of radioactivity in products of IPC degradation in lake water<sup>24)</sup>.

IPC Concentration	Days of incubation (days)	% of radioactivity in TLC plate		Total label**
		1.00-0.80* IPC	0.80-0.00 Metabolites	remaining in solution(%)
8	93	7	100	
18	89	11	100	
46	80	20	100	
400 pg/ml	0	98	2	100
	8	65	35	97
	18	56	44	82
	46	25	75	29

<sup>\*</sup> By TLC with developing solvent of chloroform + acetone (9+1)

<sup>\*\*</sup> Radioactivity in 1.0- and 10-ml portions of unextracted samples receiving 1.0  $\mu g$  and 400 pg of IPC per ml, respectively. The standard deviation for the values recorded as 100% was 2.1%.

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