

BEHAVIOR OF ^{14}C -DDT IN SUBTROPICAL FLOODED SOIL*

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ABSTRACT

A study on the behavior of p, p'-DDT in flooded soil was conducted from 1990 to 1991. ^{14}C -p,p'-DDT was used as a tracer. TLC, ARG techniques were used. A chromatography scanner was used to determine the radioactivity in spots of TLC plates. The behavior of DDT in flooded soil is different from that in upland soil. The dissipation of DDT was faster, DDT and its metabolites moved down faster, and more radioactivity became to bound residue in flooded soil as compared with those in upland situation. In extracts, DDT, DDD, DDE, DDA and an unidentified product were detected and their amounts were changed with time. About 90 % of the DDT disappeared and DDD became the key component of extractable residue within 8 weeks in flooded soil, whereas DDT took about 80 % of the methanol extractable residue in 43rd week sample of upland soil.

Keywords: DDT Dissipation Distribution Metabolites

1 INTRODUCTION

DDT [1,1,1-trichloro-2,2-bis (4-chlorophenyl) ethane] may be the archetypal persistent pesticide in the view of many scientists and the general public. Most data supporting this were generated from the temperate zone studies. However, its behavior in tropical soils was less documented^[1].

DDT was first introduced to China in 1951. According to a report, from 1951 to 1983 the accumulated production of DDT was 327 GT. During this period, 500 GT was applied for insect control. On an average 12.523 ± 1.590 kg (40 % a.i.) was applied to every hectare of cultivated land from 1952 to 1984 in China. A theoretical calculation was made that an accumulated residue of DDT was 0.3941 ± 0.0883 ppm in the cultivated layer of soil in 1980. In Zhejiang Province, the residue level of DDT would be reduced to ppb by 1990 and to ppt by 2000^[2]. DDT played a very important role with BHC in insect control in China since its introduction until the early 1980s.

Chlorinated pesticides were officially banned for production in 1983 and for use in

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1984, in China. Many factors were involved to ban the use of chlorinated pesticides. One of them was the world wide concern about their harmful residues.

It was reported that chlorinated pesticides were less persistent under high temperatures than low ones^[3,4], under flooded conditions than upland^[5,6]. Samuel et al^[7] in their study found more rapid loss of DDT in subtropical India than in temperate zones. Given that, DDT is still used in many tropical regions for the control of insect vectors of many human and animal diseases, as well as of some crop pests. Understanding its behavior in these environments is critical in the evaluation of its safety. Therefore, it would be highly worthwhile to evaluate the specificity of the behavior of DDT in the south part of China, where the climate is tropical and subtropical, and paddy rice is the most important crop.

2 MATERIALS AND METHODS

2.1 Materials and equipment

- a. ^{14}C -p,p'-DDT was supplied by IAEA (produced in the Institute of Isotopes, Hungary, Budapest);
- b. Cold p,p'-DDT, p,p'-DDE, p,p'-DDD, p,p'-DDA;
- c. The experiment site: Hangzhou is located in the $30^{\circ} 14' \text{N}$ and $120^{\circ} 10' \text{E}$;
- d. The properties of soil used in the experiment: O. M. content 3.02 %, Silt 39 %, Sand 43 %, Clay 18 %, pH value 5.9 (H_2O);
- e. Liquid scintillation counter (LSC): Tri-Carb 1990 TR;
- f. Biological oxidizer-OX 400;
- g. FJ-2109 automatic chromatography scan system;
- h. PVC cylinders: 25 cm in length \times 10 cm in diameter.

2.2 Treatment

- a. Twenty PVC cylinders were filled with 1.5 kg soil and buried into soil 1 m apart in net house under natural conditions and kept a very thin layer of water in the cylinders. Then the experiment plot was irrigated and kept flooded constantly.
- b. 630.5 kBq ^{14}C -p,p'-DDT in 0.4 ml methanol containing 1.18 mg cold p,p'-DDT was added onto water surface in each cylinder; For avoiding wash-off of radioactivity in the cylinders by heavy rain, the cylinders were 3 cm higher than the water surface on ground.
- c. The soil in cylinders was kept in flooded condition during the experiment.

2.3 Sampling and analysis

- a. Samples were taken at the 1st, 2nd, 3rd, 4th, 6th, 8th, 13th, 17th, 25th, 43rd week after treatment.
- b. The soil in cylinders was divided into 4 sections: 0—2 cm, 2—5 cm, 5—10 cm, below 10 cm. Each section was thoroughly mixed up and $20 \text{ g} \times 2$ was taken from each sample for extraction done with methanol in a Soxhlet apparatus for 6 h. Aliquots

(0.5 ml \times 2) of the concentrated extracts were determined by LSC. The remainder of the 0–2 cm extracts was purified and co-chromatographed with standards of p,p'-DDT, p,p'-DDE, p,p'-DDD, p,p'-DDA on thin layer plates coated with silicon gel HF₂₅₄. Then, the TLC was scanned by FJ-2109 TLC scanner to get spectra of p,p'-DDT and its metabolites and afterward, an AGR film was made. In order to measure the bound residue, 1.00 g of the extracted soil was combusted in the Biological Oxidizer OX-400 and the CO₂ was absorbed with LSC solution and also counted by LSC.

3 RESULTS AND DISCUSSION

3.1 The dissipation of ^{14}C -DDT in flooded soil

It is expressed in percentages of ^{14}C -residue (extractable, bound and total) over the initial ^{14}C -activity applied to each cylinder. Table 1 shows the results. The dissipation in flooded soil could be expressed by the equation $A_t = A_0 \exp(-kt)$ where A_t is ^{14}C -activity at sampling time; A_0 is ^{14}C -activity applied; k is dissipation coefficient; t is time after treatment; $k = -0.2049$ (0–6 weeks); $k = -0.0436$ (6–43 weeks).

Table 1
Dissipation of ^{14}C -DDT in paddy soil

t / week	1	2	3	4	6	8	13	17	25	43
E	68.7	55.8	36.4	35.1	26.1	26.6	28.0	25.8	19.8	25.3
B	4.5	7.0	17.9	18.5	22.3	23.5	28.3	27.7	22.0	25.6
T	73.2	62.8	54.3	53.6	48.4	50.1	56.3	53.5	41.8	50.9

E: Methanol extractable residue; B: Bound residue; T: Total residue; t: Time in Week after treatment; Errors (2σ / counts \times 100 %) for all the radioactive countings are $\leq 5\%$.

The calculated dissipation half life of p,p'-DDT in flooded soil was about 6 weeks according to the experiment results. The bound residue was increased with time and at 13th week reached the peak taking about 28 % of the total residue. The extractable residue was rapidly decreased, and within 4 weeks more than 50 % disappeared.

3.2 The distribution of ^{14}C -DDT in paddy soil

It is expressed in percentages of ^{14}C -residue in each layer (extractable, bound) over the total ^{14}C -activity at that sampling time. The results are listed in Table 2. From Table 2, it is easy to understand that most of the radioactive residue were concentrated within 2 cm depth of the column. Until the 43rd week, still about 95 % of the total residue was found within 5 cm of the soil surface. It looks DDT and its metabolites moved down quicker through soil profile in flooded situation. Radioactivity was detected at the second sampling time below 10 cm in paddy soil experiment.

3.3 The composition of DDT and its metabolites in extracts

It is expressed in percentages of radioactivity under each peak over the total radioactivity in the TLC spectra. By using TLC and ARG techniques, five spots could be recognized. Table 3 shows the relative amounts of DDA, DDD, DDT, DDE and the unidentified one. The R_f value of the unknown is 0.14.

Table 2
The Distribution of ^{14}C -DDT in paddy soil

%

L / cm	t / week	1	2	3	4	6	8	13	17	25	43
0-2	E	93.56	88.47	65.67	63.83	52.33	51.59	48.13	47.87	45.52	45.24
	B	5.77	10.48	30.97	30.9	42.54	43.99	45.49	46.35	46.41	42.39
	T	99.33	98.95	96.64	94.73	94.87	95.58	93.62	94.22	91.93	87.63
2-5	E	0.26	0.33	1.07	0.68	0.99	0.92	0.60	1.27	0.88	2.79
	B	0.38	0.67	1.69	2.15	1.88	0.91	1.49	1.88	2.17	4.46
	T	0.64	1.00	2.76	2.83	2.87	1.83	2.09	3.15	3.05	7.25
5-10	E	0.02	0.07	0.22	0.18	0.6	0.4	0.6	0.46	0.59	0.74
	B	0.04	0.03	0.33	0.86	1.38	0.77	1.86	1.68	1.86	1.8
	T	0.06	0.1	0.55	1.04	1.98	1.17	2.46	2.14	2.45	2.54
Below 10	E	0	0.01	0.02	0.02	0.1	0.1	0.44	0.54	0.41	0.9
	B	0	0	0.04	0.16	0.29	0.22	1.35	1.87	2.17	1.68
	T	0	0.01	0.06	0.18	0.39	0.32	1.79	2.41	2.58	2.58

L: Layer of the soil column; t : time in week after the treatment;

E: Methanol extractable residue; B: bound residue; T: Total residue

Table 3
The composition of ^{14}C -p,p'-DDT and its metabolites in flooded soil

%

Time	1	2	3	4	6	8	13	17	25	43
DDA	2.85	1.4	2.22	2.87	1.16	1.02	0.72	0.84	1.14	1.25
DDD	8.52	8.87	26.35	57.23	70.01	76.54	85.26	86.27	84.74	79.05
DDT	86.73	87.2	68.94	35.86	17.39	12.83	5.92	5.36	6.17	8.14
DDE	1.89	2.42	2.01	3.42	9.56	8.55	7.06	6.47	7.05	10.94
Unknown	0	0.1	0.49	0.62	1.5	1.07	1.04	1.06	0.91	0.63
T.P.A.	31.2	78.6	72.8	57.8	50.5	45.1	44.1	41.7	56.9	41.9

Time: Weeks after treatment; Unknown: Unidentified metabolite; T. P. A.: Total peak area activity (kcpm).

4 CONCLUSIONS

From the experiments conducted, the conclusions could be drawn:

a. ^{14}C -p,p'-DDT was more persistent in upland soil^[8] than in flooded soil. The dissipation half-life of ^{14}C -p,p'-DDT in upland soil was 16.6 weeks, and in flooded soil it was shorter than 6 weeks.

b. ^{14}C -DDT and/or its metabolites both in upland and flooded soils migrated to deeper layer of the soil column, ca. 95 % of the total residue kept within 0-5 cm layer of the surface soil by the end of the experiment, but p,p'-DDT and/or its metabolites was more mobile under flooded condition.

c. In the extracts, p,p'-DDT, p,p'-DDE, p,p'-DDD, p,p'-DDA and an unidentified metabolite were detected in variable amounts with time. DDT was the main component (above 80 %) at the end of upland situation, and DDD became the main metabolite (more than 50 %) of flooded soil situation within 4 weeks after the treatment.

d. The difference between upland and flooded might be caused by the soil microbial community. A report^[9] was mentioned the aerobic microbes were developed well under upland condition and the anaerobics were developed well under paddy situation.

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