

**PHOTOSENSITIZER INFLUENCE ON SOME  
ORGANOPHOSPHORUS PESTICIDES  
UNDER ULTRA VIOLET RAYS**

**M.E.A. HEGAZY<sup>2</sup>, F.A. ADAM<sup>1</sup>, SH. A. SHOKR<sup>2</sup>,  
M.F.A. SHADY<sup>1</sup>, M.EL. A.ABD BAKI<sup>1</sup>,**

*1 Faculty of Agriculture, Kafr El-Sheikh, Tanta University.*

*2 Central Agricultural Pesticides Laboratory, Agricultural Research Centre, Dokki, Egypt.*

(Manuscript received 23 November, 1997)

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**Abstract**

This investigation was carried out to study the influence of acetophenone, benzoin, benzil, p-nitrophenol and chlorophyll on the rate of Photodegradation of the active ingredients of pirimiphos-methyl, fenitrothion, malathion and prothiofos insecticides which are widely used in Egypt to protect vegetable crops from economic pests.

The results indicated that the percent loss of pirimiphos-methyl, fenitrothion, malathion, and prothiofos in methanol were 56.95, 16.40, 25 and 26.32% after 120 hours of exposure to UV-rays. The corresponding values were 50.33, 20.53, 67.57 and 50% for pirimiphos-methyl; 18.03, 19.67, 41.98 and 27.16% for fenitrothion; 22.41, 19.83, 53.06 and 26.53% for malathion; and 14.47, 17.76, 58.27, 45.27 and 42.45% for prothiofos (in methanol) when exposed to UV-rays in presence of acetophenone, p-nitrophenol, benzoin, benzil and chlorophyll for the same period of exposure, respectively. The calculated residue half lives values were 98.69, 464.4, 289.13 and 272.39 hours for pirimiphos-methyl, fenitrothion, malathion and prothiofos, while these values were 118.87, 361.99, 66.84, 65.82 and 120 hours for pirimiphos-methyl; 418.29, 379.70, 152.82, 172.43 and 262.47 hours for fenitrothion; 327.78, 376.41, 109.98, 133.05 and 269.79 hours for malathion and 532.04, 425.32, 95.17, 137.78 and 150.56 hours for prothiofos in presence of acetophenone, p-nitrophenol, benzoin, benzil, and chlorophyll, respectively.

Benzoin and benzil compounds were found to be active as photosensitizers for all the studied insecticides. Chlorophyll and acetophenone generally showed no photosensitizing effect. Only chlorophyll showed a moderate effect with fenitrothion and prothiofos insecticides. On the contrary, p-nitrophenol had photostabilizing effect on pirimiphos-methyl, malathion and prothiofos insecticides, but the effect was most evident with pirimiphos-methyl insecticide. A photostabilizing effect was clearly evident for acetophenone with prothiofos insecticide irradiated by UV-light.

## INTRODUCTION

The mechanism of photosensitization formed by the excited state charge transfer complex, by electron transfer from the singlet state of sensitizer followed by photolysis or isomerization of the ground state (charge transfer mechanism). Singlet sensitizers such as diphenyl amine were found to induce photolysis of halo-benzene while triplet sensitizers such as benzophenone did not enhance the rate of reaction (Miller and Narang, 1970). Humic acid, tryptophane and tyrosine, occurring in natural water, act as photosensitizers towards some kinds of pesticides in the environment (Draper and Crosby 1981).

This investigation aimed to study the influence of some photo-sensitizers on the rate of photodegradation of some organophosphorus pesticides irradiated by UV-rays.

## MATERIALS AND METHODS

### Pesticides used

1. Pirimiphos-methyl: (0-2-diethyl-6-methylpyrimidin-4-yl 0,0-dimethyl phosphorothioate).
2. Fenitrothion: (0,0-dimethyl 0-4-nitro-m-tolyl phosphorothioate).
3. Malathion.
4. Prothiofos: (0-2, 4-dichlorophenyl-S-propyl phosphorodithioate).

### Photosensitizers studied in this work

- a. Acetophenone (acetyl benzene).
- b. p-nitrophenol.
- c. Benzoin (benzyl phenyl carbinol) or ( $\alpha$ -hydroxy  $\alpha$ -phenyl acetophenone).
- d. Benzil (1,2 diphenyl ethane dione) or (diphenyl- $\alpha$ ,  $\beta$ -diketone).
- e. Chlorophyll.

**Procedure:** Aliquots of tested organophosphorus insecticides (5000  $\mu$ g/5ml methanol) were placed in pyrex flasks and 5 ml of photosensitizers (0.1 in methanol)

were added. Flasks were stoppered and gently shaken, then exposed to UV-rays (254 nm) at distance 12 cm. Samples of 1 ml were taken at zero time and intervals of 12, 24, 48, 72 and 120 hours.

Each sample was placed in a standard glass stoppered test tube and the solvent was evaporated to dryness and kept for determination.

**Determination:** A pye Unicam 4500 gas chromatograph equipped with a flame photometric detector operated in the phosphorus mode (526 filter) was used for determination of the tested insecticides. A pyrex glass column (1.5 m x 4 mm i.d.) was packed with 4% S.E. 30 + 6% OV 210 on gas chromosorb Q (90-100).

Temperature degrees and gas flow rates were as follows:

Column Temp. 230°C.

Detector Temp. 240°C.

Injector Temp. 235°C.

Gases flow rates were 30 ml/min. for nitrogen, hydrogen and air. Retention time for pirimiphos-methyl, fenitrothion, malathion and prothiofos under these conditions were 4.22, 5.77, 5.25 and 7.64 min., respectively. The half life time ( $t_{50}$ ) for each of the investigated insecticides was calculated using the equation of Moyer (Moyer et al. 1987).

$$t_{1/2} = \ln 2 / K' = 0.6932 / k'$$

$$K' = 1/t_{in}. \quad a/b = 1/t. \quad \ln/bx$$

Where:

$K'$  = rate of decomposition.

$a$  = Initial residue

$t_x$  = Time in hours.

$b_x$  = Residue at  $x$  time.

## RESULTS AND DISCUSSION

Effect of acetophenone, p-nitrophenol, benzion, benzil and chlorophyll as photosensitizers on the decomposition of the tested insecticides after exposure to UV - light

### 1. Pirimiphos-methyl

The data in Table 1 clearly show that the rate of decomposition of pirimiphos-methyl residues in methanol was 56.9% after 120 hours of exposure to UV-rays.

While the corresponding values were 50.33, 20.53, 68.92, 67.57 and 50% when pirimiphos-methyl in methanol exposed to UV-rays in the presence of acetophenone, p-nitrophenol, benzoin, benzil and chlorophyll for the same period of exposure, respectively. The calculated residue half life value of this insecticide was 98.69 hours, while the values were 118.87, 361.99, 66.84, 65.82 and 120 hours in the presence of acetophenone, p-nitrophenol, benzoin, benzil and chlorophyll, respectively.

## 2. Fenitrothion

The data in Table 2 represent the decomposition percentages of fenitrothion. The percent loss of fenitrothion residues in methanol was 16.40% after 120 hours of exposure to UV-rays. While the corresponding values were 18.03, 19.67, 41.98, 38.27 and 27.16% when fenitrothion in methanol was exposed to UV-rays in the presence of acetophenone, p-nitrophenol, benzoin, benzil and chlorophyll for the same period of exposure, respectively. The calculated residue half life value of this insecticide was 464.39 hours, while the values were 418.28, 379.70, 152.82, 172.43 and 262.47 hours in the presence of acetophenone, p-nitrophenol, benzoin, benzil and chlorophyll, respectively.

## 3. Malathion

The data presented in Table 3 show that the percent loss of malathion residues in methanol was 25% after 120 hours of exposure to UV-rays. While the corresponding values were 22.41, 19.83, 53.06, 46.49 and 26.53% when malathion in methanol was exposed to UV-rays in the presence of acetophenone, p-nitrophenol, benzoin, benzil and chlorophyll for the same period of exposure, respectively. The half life value of malathion was calculated at 289.13 hours, while the values were 327.78, 376.41, 109.98, 133.05 and 269.79 hours for photosensitizers mentioned above, respectively.

## 4. Prothiofos

Examination of the data in Table 4 show that the percentage loss of prothiofos residues in methanol was 26.32% after 120 hours of exposure to UV-rays. These values became 14.47, 17.76, 58.27, 45.32 and 42.45% when prothiofos in methanol was exposed to UV-rays in the presence of acetophenone, p-nitrophenol, benzoin, benzil and chlorophyll, respectively. The half life value of prothiofos was 272.39 hours which became 532.04, 425.32, 95.17, 137.78 and 150.56 hours in the pres-

Table 1. Effect of some photosensitizers on pirimiphos-methyl insecticide when exposed to UV-rays.

Time of exposure in hours	Pirimiphos-methyl		Pirimiphos-methyl + Acetophenone		Pirimiphos-methyl + p-nitrophenol		Pirimiphos-methyl + Benzoin		Pirimiphos-methyl + Benzil		Pirimiphos-methyl + Chlorophyll	
	$\mu\text{g}$ Insecticide	% Loss	$\mu\text{g}$ Insecticide	% Loss	$\mu\text{g}$ Insecticide	% Loss	$\mu\text{g}$ Insecticide	% Loss	$\mu\text{g}$ Insecticide	% Loss	$\mu\text{g}$ Insecticide	% Loss
0	500.00	00.00	500.00	00.00	500.00	00.00	500.00	00.00	500.00	00.00	500.00	00.00
12	472.10	5.58	466.00	6.80	475.10	4.98	456.09	8.78	447.39	10.52	456.09	8.78
24	432.29	13.54	448.35	10.33	461.75	7.65	371.43	25.71	390.48	21.90	416.33	16.73
48	347.38	30.52	362.00	27.60	445.35	10.93	297.96	40.41	289.79	42.04	350.00	30.00
72	310.00	38.00	345.00	31.00	414.00	17.20	225.00	55.00	210.71	57.86	292.85	41.43
120	215.23	56.95	248.34	50.33	397.35	20.53	155.41	68.92	162.16	67.57	250.00	50.00
RL-50 in hours	98.69		118.87		361.99		66.84		65.82		120.00	

Table 2. Effect of some photosensitizers on fenitrothion insecticide when exposed to UV-rays.

Time of exposure in hours	Fenitrothion		Fenitrothion + Acetophenone		Fenitrothion + p-nitrophenol		Fenitrothion + Benzoin		Fenitrothion + Benzil		Fenitrothion + Chlorophyll	
	$\mu\text{g}$ Insecticide	% Loss	$\mu\text{g}$ Insecticide	% Loss	$\mu\text{g}$ Insecticide	% Loss	$\mu\text{g}$ Insecticide	% Loss	$\mu\text{g}$ Insecticide	% Loss	$\mu\text{g}$ Insecticide	% Loss
0	500.00	00.00	500.00	00.00	500.00	00.00	500.00	00.00	500.00	00.00	500.00	00.00
12	477.77	4.45	483.33	3.33	488.88	2.22	467.94	6.41	474.68	5.06	473.41	5.32
24	465.00	7.00	455.00	9.00	470.00	6.00	428.57	14.29	422.07	15.59	454.00	9.20
48	439.00	12.20	442.30	11.54	450.00	10.00	376.62	24.68	399.32	20.14	402.59	19.48
72	426.22	14.76	431.23	13.75	434.42	13.12	333.33	33.33	346.15	30.77	378.21	24.36
120	418.00	16.40	409.83	18.03	401.63	19.67	290.12	41.98	308.64	38.27	364.19	27.16
RL-50 in hours	464.39		418.29		379.70		152.82		172.43		262.47	

Table 3. Effect of some photosensitizers on malathion insecticide when exposed to UV-rays.

Time of exposure in hours	Malathion		Malathion + Acetophenone		Malathion + p-nitrophenol		Malathion + Benzoin		Malathion + Benzil		Malathion + Chlorophyll	
	$\mu\text{g}$ Insecticide	% Loss	$\mu\text{g}$ Insecticide	% Loss	$\mu\text{g}$ Insecticide	% Loss	$\mu\text{g}$ Insecticide	% Loss	$\mu\text{g}$ Insecticide	% Loss	$\mu\text{g}$ Insecticide	% Loss
0	500.00	00.00	500.00	00.00	500.00	00.00	500.00	00.00	500.00	00.00	500.00	00.00
12	477.26	4.55	477.26	4.55	488.63	2.27	460.31	7.94	452.37	9.53	484.12	3.18
24	468.75	6.25	469.45	6.11	471.69	5.66	416.66	16.67	434.02	13.20	454.86	9.03
48	436.36	12.73	431.81	13.64	440.91	11.82	332.14	33.57	403.57	19.29	420.60	15.88
72	410.44	17.91	406.71	18.66	421.64	15.67	291.66	41.67	329.86	34.03	406.25	18.75
120	374.99	25.00	387.93	22.41	400.86	19.83	234.68	53.06	267.57	46.49	367.34	26.53
RI-50 in hours	289.13		327.78		376.41		109.98		133.05		269.79	

ence of photosensitizers mentioned above before, respectively.

Benzoin and benzil compounds were found to be active as photosensitizers for all the studied insecticides. Chlorophyll and acetophenone generally showed no photosensitizing effect. Only chlorophyll showed a moderate effect in both fenitrothion and prothiofos insecticides.

On the contrary, *p*-nitrophenol had photostabilizing effect with primiphos-methyl, malathion and prothiofos, but the effect was most evident with pirimiphos-methyl insecticide. The photostability effect was clearly evident for acetophenone with prothiofos insecticide irradiated by UV-light.

Photolysis is one of the various factors determining the fate of a pesticide in the environment. A number of recent studies have examined the photolysis of pesticides in water and air as well as on plant and soil surfaces by sunlight and ultraviolet light irradiation (Draper and Crosby, 1981; Wong and Crosby, 1981). Light plays an important role in the behaviour of pesticides in the environment, when the molecule which absorbs a photon becomes unstable, and undergoes a variety of competing primary processes, such as chemical reaction isomerization etc., to return to a stable state (Zepp and Cline, 1977). Photosensitization occurs by the formation of the excited state charge transfer complex, by electron transfer from the singlet state of sensitizer followed by photolysis or isomerization of the ground state (charge transfer mechanism). Singlet sensitizers such as diphenyl amine have been found to induce photolysis of halobenzene, while triplet sensitizers such as benzophenone do not enhance the rate of reaction (Miller and Narage, 1970). Using photosensitizers such as acetone, riboflavine, rhodamine B, chlorophyll and methylene blue results in the rapid disappearance of ETU under both natural and artificial light (Ross and Crosby, 1973). Photosensitization via energy transfer and/or photoinduced degradation of pesticides is known to occur in the presence of humic substance (Coudry, 1982).

The triplet photosensitizer, acetone, together with naturally occurring substances in river and sea water and humic acid were found by Takahashi *et al.* (1985). It has been reported that humic acid, tryptophane and tyrosine, occurring in natural water, act as photosensitizers towards some kinds of pesticides in the environment (Draper and Crosby, 1981). The excited triplet state of humic substances can transfer its energy to chemical adsorbed, but that its excited singlet plays an insignificant role due to its extremely short life time (Zepp *et al.*, 1985). One can



conclude that both photosensitizing agents (e.g., humic acids or some pigments that can act in this way in nature) and iron compounds, usually occurring at levels of the order of 0.1 mg/l in surface water, can play a significant role in kinetics of photo-oxidative degradation of pesticides (Sedlak et al., 1992). It has been reported also that the soil transition metal catalysis and a change in the adsorption spectra of chemical in sorbed states resulted in acceleration of the photolysis (Plimmer 1972). However the photostability of pesticides is due to the lower quantum yield and lower rate of specific sunlight absorption (Takahashi et al. 1985). The efficiency of the photostabilization affected by several factors. Adsorption to clays containing a minimal concentration of lattice iron was found to slow down the photo inactivation, probably due to charge-transfer processes occurring between the insecticide molecules and the  $Fe^{3+}$  ions. When the cationic dye 3,6-diamino-10-methylcrininium (AF) was co-adsorbed intermolecular interaction between it and the pesticide resulted in a considerable improvement of the photostabilization yield (Rozen and Margulies 1991). The photodegradation of parathion and chlormphos insecticides can be sensitized by a number of compounds which are present in biochemical systems such as pyruvic and  $\alpha$ -ketoglutaric acid (Buckland and Davidson 1987), 2,4,5-trichlorophenoxy acetic acid in water was photolyzed by artificial UV-light, the rate of reaction increased twice in the presence of acetone or riboflavine as photosensitizer (Crosby and Wong, 1973).

Table 4. Effect of some photosensitizers on prothiofos insecticide when exposed to UV-rays.

Time of exposure in hours	Prothiofos		Prothiofos + Acetophenone		Prothiofos + p-nitrophenol		Prothiofos + Benzoin		Prothiofos + Benzil		Prothiofos + Chlorophyll	
	$\mu\text{g}$ Insecticide	% Loss	$\mu\text{g}$ Insecticide	% Loss	$\mu\text{g}$ Insecticide	% Loss	$\mu\text{g}$ Insecticide	% Loss	$\mu\text{g}$ Insecticide	% Loss	$\mu\text{g}$ Insecticide	% Loss
0	500.00	00.00	500.00	00.00	500.00	00.00	500.00	00.00	500.00	00.00	500.00	00.00
12	481.26	3.75	482.26	3.55	490.69	1.86	460.53	7.89	4488.89	2.22	487.80	2.44
24	459.14	8.17	465.52	6.89	474.14	5.17	416.67	16.67	476.19	4.76	464.29	7.14
48	430.95	13.81	445.67	10.87	457.03	8.59	344.44	31.11	411.11	17.78	422.22	15.56
72	390.15	21.97	439.07	12.19	431.18	13.76	307.69	38.46	375.00	25.00	379.81	24.04
120	368.42	26.32	427.63	14.47	411.18	17.76	208.63	58.27	273.38	45.32	287.76	42.45
RL <sub>50</sub> in hours	272.39		532.04		425.32		95.17		137.78		150.56	

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## تأثير المحسسات الضوئية علي بعض المبيدات الفوسفورية المعرضة للأشعة فوق البنفسجية

محمد السعيد علي حجازي<sup>١</sup>، فكري أحمد آدم<sup>٢</sup>، شكر عبد السلام شكر<sup>١</sup>،

محمد فاضل عبد الحليم شادي<sup>٢</sup>، محمد عبد السلام عبد الباقي<sup>٢</sup>

١ العمل المركزي للمبيدات - مركز البحوث الزراعية - الدقي - جيزة.

٢ كلية الزراعة بكفر الشيخ - جامعة طنطا.

تمت دراسة تأثير الأسيوتوفينون والبارانيتروفينول والبنزوين والبنزويل والكلوروفيل كمحسسات ضوئية عند تعرض متبقيات مبيدات البيريثيفوس - ميثيل والفنتروثيون والملاثيون والبروثيوفوس للأشعة فوق البنفسجية. وقد أوضحت الدراسة مايلي:

- عند تعريض متبقيات البيريثيفوس-ميثيل والفنتروثيون والملاثيون والبروثيوفوس في الميثانول للأشعة فوق البنفسجية لمدة ١٢ ساعة كانت النسب المئوية للفاقد في متبقياتها هي ٩٥، ٩٥، ٤٠، ٤٠، ١٦، ٢٢، ٢٢، ٢٢٪ علي التوالي. وكانت فترات نصف العمر لمتبقيات هذه المبيدات هي ٦٩، ٩٨، ٢٩، ٤٦٤، ١٢، ٢٨٩، ٢٩، ٢٧٢ ساعة علي نفس الترتيب. وقد اختلفت معدلات التحطم الضوئي عند تعريضها للأشعة فوق البنفسجية في وجود المحسسات الضوئية: الأسيوتوفينون والبارانيتروفينول والبنزوين والبنزويل والكلوروفيل حيث كان لهذه المحسسات الضوئية تأثيرات مختلفة علي معدلات التحطم الضوئي وعلي فترات نصف العمر لمتبقيات هذه المبيدات.

- كان لوجود المحسسات الضوئية مع متبقيات مبيد البيريثيفوس تأثيرات متباينة فقد أدى الأسيوتوفينون والنيتروفينول الي خفض نسبة الفقد في متبقياته عند تعريضها للأشعة فوق البنفسجية بينما زاد معدل تحطم المبيد في وجود البنزوين، البنزويل والكلوروفيل.

- أما بالنسبة لمبيد الفنتروثيون كان لجميع المحسسات الضوئية المستخدمة تأثيرا إيجابيا علي تحطم المبيد بدرجات متفاوتة، فعند تعريض متبقيات المبيد في الميثانول للأشعة فوق البنفسجية لمدة ١٢ ساعة كانت نسبة الفقد فيها ١٦، ٤٪ بينما كانت نسب الفقد في المبيد عند التعرض في وجود الأسيوتوفينون والبارانيتروفينول والبنزوين والبنزويل والكلوروفيل هي ١٨، ٠٢، ٦٧، ١٩، ٩٨، ٤١، ٢٧، ٢٨، ١٦، ٢٧٪ علي الترتيب مما كان له أثر في خفض فترة نصف العمر لمتبقيات هذا المبيد في وجود المحسسات الضوئية.

- أما مبيد الملاثيون فقد كان للأسيوتوفينون والبارانيتروفينول تأثيرا مخفضا لمعدل تحطمه الضوئي بالأشعة فوق البنفسجية وبالتالي الي زيادة في فترة نصف العمر للمبيد، وعلي العكس من ذلك أدى استخدام البنزوين والبنزويل والكلوروفيل الي زيادة معدلات التحطم الضوئي للملاثيون وخفض فترة نصف العمر لمتبقيات المبيد.

- كما أوضحت الدراسة أن استخدام الأسيوتوفينون والبارانيتروفينول أدت الي خفض معدل التحطم الضوئي للبروثيوفوس بالأشعة فوق البنفسجية وبالتالي زيادة فترة نصف عمر متبقياته، اما البنزوين والبنزويل والكلوروفيل فكان لهم تأثيرا واضحا في زيادة معدلات تحطم المبيد وخفض نصف العمر لمتبقياته، مما يؤكد أن المحسسات الضوئية المستخدمة تلعب دورا في عمليات تحطم المبيدات سواء بالسلب أو الإيجاب.