THERMAL AND PHOTODECOMPOSITION OF SOME ORGANOPHOSPHORUS INSECTICIDES

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Abstract

This investigation was carried out to study the influence of different temperature degrees, short ultra -violet and direct sun light on the rate of decomposition of pirimphos - methyl, fenitrothion ,malathion and prothiofos insecticides, which are widely used in Egypt to protect vegetable crops from economic pests.

A. Stability of pirimphos-methyl, fenitrothion, malathion and prothiofos insecticides under different temperature degrees .

The results indicated that the increase of temperature degrees means an increase in insecticide residue loss. the percentage loss of pirimphos-methyl, fenitrothion, malathion and prothiofos insecticides was 8.69%,1.33%, 2.42% and 0.57% at20°C, 41.70% , 23.13%, 20.23% and12.86% at 30°C ;97.56%, 60.28% ,73.56, and 55.81% at 40°C and 99.98%, 97.38%, 93.855% and 89.59% at 50°C, respectively after 192 hours of exposure.

On the other hand, the half life values of these insecticides were 1463.68, 9918.52, 5432.99 and 23243.03 hours at 20°C ; 246.68, 506.08, 587.26 and 966.71 hours at 30°C; 61.51, 134.67, 123.22 and 162.97 hours at 40°C; and 18.45, 40.47, 56.49 and 93.18 hours at 50°C, respectively.

In general, icreasing temperature degrees and prolongation of exposure increase the percentage loss of insecticide. Prothiofos insecticide decomposed less than the three others. Fenitrothion, and malathion showed moderate thermal degradation, pirimphos-methyl had the highest thermal decomposition during the periods of experiment.

B.Ultra violet ray effect.

The obtained results showed that fenitrothion, malathion and prothiofos residues were more stable than pirimphos-methyl when exposed to UV-rays. The percentage loss of pirimphos-methyl, fenitrothion, malathion, and prothiofos insecticides after 12 hours were 78.97%, 39.46%, 42.47% and 48.30% respectively. The half life values of these insecticides were 6.41, 14.96, 13.55, and 12.61 hours, respectively.

The rate of degradation of the four tested insecticides varied according to their chemical structure, time of exposure and the wavelengh of UV-rays used. Pirimphos-methyl was the most affected by UV irradiation among the four insecticides.

C. Sunlight effect .

The tested insecticides greatly deteriorated when exposed to direct sunlight. The percentage loss of pirimphos-methyl, fenitrothion, malathion and prothiofos insecticides were 100%, 97.45%, 97.67% and 95.85%, respectively after 48 hours of exposure. The half life values of these insecticides were 1.23, 4.39, 4.74 and 7.99 hours, respectively.

It can be concluded that the active ingredent of the four tested insecticides greatly deteriorate when exposed to direct sunlight. Pirimphos-methyl insecticide showed highly significant degradation compared with the other tested insecticides. On contrary, prothiofos insecticide showed more persistence when exposed to direct sulight, followed by fenitrothion and malathion. Sunlight is found to be more effective than UV-rays in accelerating the photodecomposition of the insecticides, this may be due to thermal evaporation and light intensity considerations.

INTRODUCTION

In Egypt, pesticides are applied to control economic pests during the summer season where the temperature is almost always high because of the long sunny clear days. the ultraviolet component of sunlight which varied from 240 to 400 nm is responsible for pesticide photolysis in the environment. Both heat and light might affect the efficiency of pesticides which is measured by the duration of their residual effect.

The present study was undertaken to investigate the effect of temperature, UV-rays (short waves) and direct sunlight on the stability and degradation of the active ingredient of pirmphos-methyl, fenitrothion, malathion, and prothiofos insecticides.

MATERIALS AND METHODS

Pesticides used

1. Pirimphos - methyl: (O-2 diethyl- 6- methylpyrimidin-4yl O,Odimethyl phosphorothioate).

2. Fenitrothion: (O.O-dimethyl O- 4- nitro-m-tolyl phosphorothioate).

3.Malathion S-1,2- bis (ethoxycardonyl) ethyl O,O-dimethyl phosphorodithioate.

4. Prothiofos: (O-2,4- dichlorphenyl-S- propyl phosphorodithioate).

Procedure

Aliquots of the tested organophosphorous insecticides representing one milliter

ethyl acetate containing 1000 ug a.i. were each spread as uniformly as possilble on the surface of uncovered petri dishes (5cm i.d.)

The organic solvent (ethyl acetate) was left to dry at room temperature and the resulting deposits were divided and subjected to different treatments. The 1st set of treatment petri- dishes were exposed to 20,30,40, and 50°C for 4,12,24,48,96 and 192 hours inside a dark electric oven provided with a temperature regulating system. The 2nd set was exposed to short waves of an ultraviolet lamp (254nm) at a distance of 12 cm for 1,2,4,8and 12hours. The 3rd set was exposed to normal and direct sunlight regime for 1,4,12,24,48,72 and 96 hours, maximum temperature ranged between 35 and 37°C.

The residues of tesed insectides which remained on exposed surfaces were quantitatively transferrd to standard glass stopper test tubes tubes with ethyl acetate after which the solvent was evaporated under reducd pressure to dryness and the residues were ready for determination by gas liquid chromatography.

Determination

A pye Unicam 4500 gas chromatograph equipped with a flame photometric detector operated the phosphorus mode (526 filter) was used for determination of the tested insecticides. A pyrx glass column (1.5m X4mm i.d.) was packed with 4% S.E.- 30+6%OV- 210 on gas chromosorb Q (90-100).

Temp. 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 times for pirimphos- methyl, fenitrothion, malathion and prothiofos under these condition were 4.22,5.77,5.25 and 7.64 min., respectively. The half life time(t1/2) for each of the investigated insecticides was calculated using the equation of Moye (Moye *et al.* 1987).

$$t1/2 = \frac{\ln 2}{K`} = \frac{0.6932}{K`}$$

$$K` = \frac{1}{t_x} \ln \frac{a}{b_x}$$

where:

K` = rate of decomposition. t_x = time in days

a = initial residue. b_x =residue at x time

RESULTS AND DISCUSSION

A. Effect of temperature degrees on the insecticides.

1. Pirimphos - methyl insecticide

Examination of the data in table 1 clearly showed that th rate of the persistence of pirimphos- methyl on glass surfaces was influenced by temperature and the period of exposure.

The percent loss of pirimphos-methyl residues was 9.25, 23.84, 36.30, 60.14,85.00,99.00 and 99.98% after 1,4,12,24,48,96 and 192 hours of exposure at 50°C, respectively. These values were 2.60,6.86,10.72,23.75,41.78, 70.18,97.56% and 0.27, 3.56, 5.79, 9.59, 14.67, 20.37, 41.70,% when pirimphos- methyl was exposed to 40, and 30°C for the same period of exposure, respectively. The data also showed that there no loss of pirimphos-methyl till 48 hours of exposure at 20°C, while the percent loss was 1.01 and 8.69% after 96 and 192 hours of exposure at 20°C, respectively. The calculated half life values of this insecticide were 1463.68, 246.68, 61.51and18.45 hours at 20,30,40, and 50°C, respetively.

2. Fenitrothion insecticide

Data in table 2 clearly showed that the rate of persistence of fenitrothion on glass surface was influenced by temperature and the period of exposure.

There was an evident positive relationship between temperature and insecticide degradation. The percent loss of fenitrothion residues was 4.07, 12.20, 20.00, 33.71,53.18,75.08 and 97.37% after1,4, 12, 24, 48, 96 and 192 hours of exposure at 50°C, respectively. The corresponding values when fenitrothion was exposed to 40 and, 30°C for the same periods were 1.32, 3.93, 11.27, 16.19, 22.30, 38.99,60.28%, and 00.00,1.78, 4.09, 7.41,10.40, 17.31 and 23.13 % respectively. The data also showed that there was no loss of fenitrothion till 97 hours from exposure at 20°C, while the percent loss was 1.33% after 192 hours of exposure at

Table 2. Effect of different temperatures on fenitrothion insecticide.

Time of exposure in hours	20℃		30℃		40℃		50℃	
	μg Insecticide	% Loss	μg Insecticide	%Loss	μg Insecticide	%Loss	μg Insecticide	%Loss
0	1000.00	00.00	1000.00	00.00	1000.00	00.00	1000.00	00.00
1	1000.00	00.00	1000.00	00.00	986.85	1.32	959.35	4.07
4	1000.00	00.00	982.22	1.78	960.67	3.93	878.05	12.20
12	1000.00	00.00	959.11	4.09	887.33	11.27	800.00	20.00
24	1000.00	00.00	925.83	7.41	838.10	16.19	662.95	33.71
48	1000.00	00.00	896.00	10.40	776.98	22.30	468.20	53.18
96	1000.00	00.00	826.88	17.31	610.09	38.99	249.18	75.08
192	986.67	1.33	768.75	23.13	397.18	60.28	26.35	97.37
RL_{50} in hours	9918.02		506.08		134.67		40.47	

20°C. The calculated half life values of this inseticide were 9918.02, 506.08, 134.67 and 40.47 hours at 20,30,40and 50°C, respectively.

3. Malathion insecticide

The data in table 3 clearly showed that the rate of persistence of malathion on glass surface was influenced by temperature and the period of exposure.

There was an evident positive relationship between temperature and insecticide degradation. The percent loss of malathion residues was 3.20, 13.95, 19.01, 30.00, 44.51, 74.74 and 93.85% after 1, 4, 12, 24, 48, 96 and 192 hours of exposure at 50°C, respectively. The corresponding values when malathion was exposed to 40, and 30°C for the same periods were 1.62, 4.86, 11.42, 17.25, 26.08, 71.73, 73.56 and 00.00, 0.51, 2.29, 3.94, 7.67, 12.35 and 20.28%, respectively. The data also showed that there was no loss of malathion till 96 hours of exposure at 20°C, while the percent loss was 2.42% after 192 hours of exposure at 20°C. The calculated half life values of this insecticide were 5432.99, 587.26, 123.22 and 56.49 hours at 20,30, 40 and 50°C, respectively.

4. Prothiofos insecticide

Examination of the data in table 4 clearly showed that the rate of persistence of prothiofos on a glass surface was influenced by temperature and the period of exposure. The positive relationship between temperature and the degradation process was noticed . The percent loss of prothiofos residues was 1.68, 5.19, 9.07, 19.00, 30.03, 62.90 and 89.50 % after1,4, 12,24, 48,96 and 192hours of exposure at 50°C, respectively. The corresponding values when prothiofos was exposd to 40 and, 30°C for the same periods were 0.96 3.47, 7.86, 12.96, 20.98, 38.25, 55.81 % and 00.00,00.00, 00.00, 0.82,3.24, 6.85, 9.07 and 12.86%, respectively. The data also showed that there was no loss of prothiofos till 96 hours of exposure at 20°C, while the percent loss was 0.57 % after 192 hours of exposure at 20°C . The calculated half life values of this insecticide were 23243.03, 966.71, 162.97 and 93.18 hours at 20, 30, 40 and 50°C, respectively.

The results clealy showed that rate of persistence of the four tested insecticides were influenced dy many factors, including chemical structure, vapour pressure concentration of insecticide applied, temperature degrees and the period of exposure. In general, increasing temperature degrees increased the rate of residues degradation. Prothiofos insecticide had the least loss after 24hours compared with

Table 3. Effect of different temperatures on malathion insecticide.

Time of exposure in hours	*20℃*		*30℃*		*40℃*		*50℃*	
	μg Insecticide	*% Loss*	*μg Insecticide*	*%Loss*	*μg Insecticide*	*%Loss*	*μg Insecticide*	*%Loss*
0	1000.00	00.00	1000.00	00.00	1000.00	00.00	1000.00	00.00
1	1000.00	00.00	1000.00	00.00	983.79	1.62	968.00	3.20
4	1000.00	00.00	994.91	0.51	951.43	4.86	860.47	13.95
12	1000.00	00.00	977.13	2.29	885.83	11.42	809.90	19.01
24	1000.00	00.00	960.60	3.94	827.55	17.25	700.00	30.00
48	1000.00	00.00	923.27	7.67	739.21	26.08	554.87	44.51
96	1000.00	00.00	876.49	12.35	582.71	41.73	252.63	74.74
192	975.80	2.42	797.21	20.28	264.41	73.56	61.54	93.85
RL_{50} in hours	5432.99		587.26		123.22		56.49	

Table 4. Effect of different temperatures on prothiofos insecticide.

Time of exposure in hours	20℃		30℃		40℃		50℃	
	µg Insecticide	% Loss	µg Insecticide	%Loss	µg Insecticide	%Loss	µg Insecticide	%Loss
0	1000.00	00.00	1000.00	00.00	1000.00	00.00	1000.00	00.00
1	1000.00	00.00	1000.00	00.00	990.45	0.96	983.17	1.68
4	1000.00	00.00	1000.00	00.00	965.31	3.47	948.15	5.19
12	1000.00	00.00	991.81	0.82	921.41	7.86	909.33	9.07
24	1000.00	00.00	967.65	3.24	870.36	12.96	810.05	19.00
48	1000.00	00.00	931.48	6.85	790.20	20.98	699.71	30.03
96	1000.00	00.00	909.33	9.07	617.54	38.25	370.97	62.90
192	994.29	0.57	871.38	12.86	441.90	55.81	104.96	89.50
RL_{50} in hours	23243.03		966.71		162.97		93.18	

the other three insecticides. The percentage loss was 19.00% compared with 30.00, 33.71 and 60.14% for malathion, fenitrothion and pirimphos-methyl, respectively. It is clear that the percent loss of insecticide residue gradually increased with prolongation of the exposure period.

From the practical point of view in the pest control programme, it can be recommended to use prothiofos in area of dominant high temperature. Moreover, this clearly showed that the interval between successive sprays should be shorter at high temperature and vice versa.

Fenitrothion and malathion showed moderate degradation when exposed to different degrees of temperature within the period of experiment. The percent loss steadly increased by time till 192 hours (8 days). It is clear that the percent loss of the two insecticides residues gradually increase with the prolongation of the exposure period and this was pronounced at all temperatures.

Pirimiphos-methyl showed a high degradation rate when exposed to high degrees of temperature (40 and 50^{o}C) within the period of experiment. So, it is recommended for use in areas of dominant low temperatures 20-30^{o}C during the winter season.

Several investigators had studied and comfirmed the role of temperature in degradation of insecticides (Tantawy and Hussein, 1978; Hegazy *et al.*, 1982 and 1987; Khalil *et al.*, 1984a and b; Abu - zahw *et al.*, 1988).

B. Effect of UV - rays on the insecticides

Data in table 5 showed that the decomposition percentages of pirimphos- methyl, fenitrothion, and prothiofos, on glass surfaces increased gradually after exposure to UV-rays.

The percent of loss for pirimphos- methyl, fenitrothion, malathion and prothiofos were 10.02, 4.04, 5.37 and 3.83% after one hour exposure to UV-rays. The decomposition percentages of pirimphos- methyl increased to 20.36, 35.12, 69.02 and 78.97% after 2,4 and 12 hours of exposure to UV-rays, respectively. While these values were 8.70, 18.77, 25.41, 39.46%; 10.28, 20.54, 36.13, 42.47%and 17.23, 22.58, 33.79, 48.30% when fenitrothion, malathion, and prothiofos were exposed to UV-rays for the same periods of exposure, respectively.

The residue half life values were 6.41,14.96,13.55 and 12.61 hours for pirimphos- methyl, fenitrothion, malathion, and prothiofos, respectively.

The results clearly showed the rate of degradation of four tested insecticides varied according to their chemical structure, time of exposure and wavelength of UV-rays used. Pirimiphos-methyl was the most affected by UV irradiation among the four insecticides. Residues were considerably degraded at different rates due to the chemical nature of each compound and the prolongation of exposure time.

Generally, it is found that photodegradation is positively correlated with the exposure period. These results are in accordance to great extent with those obtained by several investigators (Riskalla, 1975; Greenhalgh and Marshall 1976; El-sayed *et al.*, 1980; Abdel-Razik *et al.*, 1982; Hegazy *et al.*, 1982 and 1987; Abu-Zahw *et al.*, 1988.; Chkuwudebe *et al.* 1989).

The effect of ultraviolet (UV) light on pesticides is of considerable interest to the research workers of pesticides. It has been demonstrated that UV light induced chemicals changes on large number of pesticides.

As to organophosphorus pesticides, several types of photodecomposition such as hydrolysis, oxidation and isomerization. If similar reactions occur under field condition, such an investigation is of great importance in view of environmental contamination, pesticide residues in agricultural products and practical use of pesticides (murai and Igawa 1977).

C. Effect of sunlight on the insecticides

Data presented in table 6 show that the percentage of loss for pirimiphos - methyl, fenitrothion, malathion and prothiofos were 45.18, 5.48, 6.13 and 0.82%, respectively after one hour of expoure to direct sunlight. The decomposition percentages of pirimiphos-methyl rapidly increased to 89.58,97.79,98.52,100,100and 100% after 4,12,24,48,72 and 96 hours, respectively of exposure to direct sunlight. While these values were 46.84, 66.24, 90.70,97.45, 99.40,99.96%; 44.26,67.17,91.09,97.67, 99.80, 99.97% and 29.32, 57.15, 86.77, 95.85, 98.31, 99.93% when fenitrothion, malathion and prothiofos were exposed to direct sunlight for the same periods of exposure, respectively.

The residue half life values were 1.23, 4.39, 4.74 and 7.99 hours for pirimphos - methyl, fenitrothion, malathion and prothiofos, respectively.

It is concluded that the residues of the tested insecticides are greatly deterioratd when exposed to direct sunlight especially for longer periods. Pirimiphos - methyi insecticide showed significant degradation when compared with the other three insecticides. On contrary, prothiofos showed more persistence when exposed to direct sunlight followed by fenitrothion and malathion. The results obtained agreed with the findings of Marei *et al* (1969), El- sayed *et al* (1980), Abdel- Razik *et al* (1982), khalil (1984-a and b), Hegazy *et al* (1987), Abu- zahw *et al* (1988), and Chkuwudebe *et al* (1989)

Considering the time of exposure to sulight, it is obvious that 24 hours exposure seemed to be the critical interval in determining the degradation percent. Also, it can be observed that sulight is found to be more effective than UV-rays in accelerating the photodecomposition of pirimiphos-methyl, fenitrothion, malathion and prothiofos, this may due to thermal, evaporational and light intensity considerations.

Table 5. Effect of UV-rays on organophosphorus insecticides.

Time of exposure in hours	*Pirimiphos- methyl*		*Fenitrothion*		*Malathion*		*Prothiofos*	
	µg Insecticide	*% Loss*	*µg Insecticide*	*%Loss*	*µg Insecticide*	*%Loss*	*µg Insecticide*	*%Loss*
0	1000.00	00.00	1000.00	00.00	1000.00	00.00	1000.00	00.00
1	899.82	10.02	959.60	4.04	946.26	5.37	961.74	3.83
2	796.43	20.36	912.99	8.70	897.20	10.28	827.74	17.23
4	648.78	35.12	812.31	18.77	794.63	20.54	774.22	22.58
8	309.84	69.02	745.86	25.41	638.71	36.13	662.05	33.79
12	210.30	78.97	605.38	39.46	575.27	42.47	516.99	48.30
RL_{50} in hours	6.41		14.96		13.55		12.61	

Table 6. Effect of sunlight on organophosphorus insecticides.

Time of exposure in hours	Pirimiphos- methyl		Fenitrothion		Malathion		Prothiofos	
	µg Insecticide	% Loss	µg Insecticide	%Loss	µg Insecticide	%Loss	µg Insecticide	%Loss
0	1000.00	00.00	1000.00	00.00	1000.00	00.00	1000.00	00.00
1	548.20	45.18	945.20	5.48	938.67	6.13	991.78	0.82
4	104.19	89.58	531.61	46.84	557.42	44.26	706.81	29.32
12	22.14	97.79	337.63	66.24	328.35	67.17	428.51	57.15
24	14.77	98.52	93.01	90.70	89.06	91.09	132.31	86.77
48	UND	100.00	25.50	97.45	23.29	97.67	41.48	95.85
72	UND	100.00	6.04	99.40	2.05	99.80	16.89	98.31
96	UND	100.00	0.38	99.96	0.35	99.97	0.66	99.93
RL_{50} hours	1.23		4.39		4.74		7.99	

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التحطم الحراري والضوئي لبعض المبيدات الفوسفورية العضوية

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ا- تاثير درجات الحراره المختلفه على تحطم متبقيات البيريموفوس - ميثيل والفنتروثيون والملاثيون والبرو ثيوفوس .

اوضحت النتائج انه عند تعويض متبقيات البيريموفوس - ميثيل والفنتروثيون والملاثيون والبروثيوفوس لدرجات حرارة مختلفة كانت النسبة المئوية للفاقد بعد ١٩٢ ساعة هي ٨,٧٩٪ و ١,٣٣٪ و٢,٤٢ ٪ و ٠,٥٧ ٪ عند ٥٢٠ م . وكانت ٤١,٧٠ ٪ و ٢٣,١٣ ٪ و ٢٠,٢٨ ٪ و ١٢,٨٦ ٪ و عند ٥٣٠م . ايضـا كـانت ٩٧,٥٦ ٪ و ٦٠,٢٨ ٪ و ٧٣,٥٦ ٪ و ٥٥,٨١٪ عند ٥٤٠ م . وكـانت ٩٩,٩٨ ٪ و ٩٧,٣٨ ٪ و ٩٣,٨٥ ٪ و ٨٩,٥٠ ٪ عنـد ٥٥٠ م . علي التوالى وكانت فـترات نصف العمـر هي ١٤٦٣,٧٨ ، ٩٩١٨,٥٢ ٢٣٢٤٣,٠٣٠٥٤٣٢,٩٩ ساعـة عند ٥٢٠ م. وكـانت ٢٤٦,٦٨ و ٥٠٦,٠٨ و ٥٨٧,٢٦ و ٩٦٦,٧١ سـاعـة عند ٥٣٠م . و ٦١,٥١و ١٣٤,٦٧ و١٣٢,٢٢ و ١٦٢,٩٧ سـاعـة عند ٥٤٠م . وايضـا كـانت ١٨,٤٥و ٤٨ ، ٤٠ و ٥٦,٤٩ و ٩٣,١٨ ساعة عند ٥٥٠ م على التوالى .

ب - تأثير الاشعة فوق البنفسجيه على تحطم متبقيات البيريموفوس - ميثيل والفنتروثيون والملاثيون والبروثيوفوس .

اوضحت النتائج انه عند تعرض متبقيات البيريموفوس - ميثيل و الفنتروثيون والملاثيون و البروثيوفوس للاشعة فوق البنفسجيه كانت النسبة المئوية للفاقد بعد ١٢ ساعة هى ٩٧ ، ٨٧ ٪ و ٤٦، ٣٩ ٪ و٤٧ ، ٤٢ ٪ و٣٠ ، ٤٨ ٪ على التوالى وكانت فترات نصف العمر هى ٤١ ، ٦ و ٩٧ ، ١٤و ٥٥ ,١٣ و ٦١ ، ١٢ ساعة على التوالى .

ب - تأثير ضوء الشمس المباشر على تحطم متبقيات البيريموفوس - ميثيل والفنتروثيون والملاثيون والبروثيوفوس .

اوضحت النتائج انه عند تعريض متبقيات البيريموفوس - ميثيل والفنتروثيون والملاثيون و البروثيوفوس لضوء الشمس المباشر كانت النسبة المئويه للفاقد بعد ٤٨ ساعة ..١ ٪ و ٩٧,٤٥و ٩٧,٦٧٪ و ٨٥ ، ٩٥ ٪ على التوالى وكانت فترات نصف العمر هى ٢٣ ، ١ و ٤,٣٩ و ٤,٧٤ و ٩٩ , ٧ ، ساعة على التوالى .