Pakistan J. Sci. Ind. Res., Vol. 16, No. 5, October 1973

SCREENING TESTS OF SUBSTITUTED PHOSPHINE OXIDES, PHOSPHONIC AMIDES, METHYLMELAMINES AND DIAMINOTRIAZINES FOR STERILIZATION OF LABORATORY-REARED STRAIN OF HOUSEFLIES, MUSCA DOMESTICA (L.)

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(Received April 28, 1973; revised August 10, 1973)

Abstract. Toxicological studies of phosphine oxides, phosphonic amides, methylmelamines and diaminotriazines could not be correlated with their sterilizing effect. Phosphine oxides inhibited oviposition at 0.5% concentration. At 0.25% dose the fecundity was appreciably decreased and the sterility induced ranged between 53–100%. Phosphonic amides proved much active as compared to phosphine oxides. Methylmelamines and diaminotriazines were less toxic as well as less effective than phosphine oxides and phosphonic amides.

Substituted phosphine oxides and phosphonic amides indicated no sex specificity while methylmelamines seemed to effect male flies slightly more than the females. Diaminotriazines on the other hand proved more effective in females.

The derivatives of aziridines are the most important group of chemosterilants; and most of the work on structure-activity relationship and metabolism in insects has been done with aziridinyl compounds.[1–3] Other data on chemosterilant activity of phosphine oxides and aziridines were reported by Borkovec *et al.*[4] Screening tests of some of the aziridines and triazines were done by Fye *et al.*[5,6] while Chang and Borkovec[7] reported the structure-activity relationship in analogues of Tepa and Hempa. Chang *et al.*[8] also reported the sterilizing activity of phosphine oxides and sulphides in male houseflies. LaBrecque *et al.*[9] observed melamines as active chemosterilants for houseflies. The results of experiments with active triazines and melamines were published by Borkovec and Terry,[10] Borkovec and DeMilo[11] and LaBrecque *et al.*[9] The recent work on triazines is that of Borkovec *et al.*[12] Matolin and Landa[13] studied the physiological and cytological effects and metabolism of furyltriazines.

The present paper encompasses the results of 5 substituted bis(1-aziridinyl)phosphine oxides (alkoxyphosphine oxides), 7 substituted methylmelamines, 6 phosphonic amides (alkylaminophosphine oxides) 7 diaminotriazines with their sterilization capacity compared with standard chemosterilants (Tepa, Hempa and Hemel). Some of the work on such compounds has been accomplished either on mixed sexes or on male flies only. In the present study stress has been given on sex specificity of these compounds against P.C.S.I.R.-strain of houseflies.

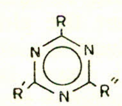
As the physical and chemical characteristic of chemosterilants is known to vary under different experimental conditions, methods of applications, stage of development of insects, concentration, treatment time and the strain of the species used. By keeping the above facts in view the present study also includes the data on the mortality, hatchability of eggs and per cent pupation induced by these compounds.

Materials and Methods

Candidate Chemosterilants. The compounds in the first category are substituted bis(1-aziridinyl)-phosphine oxides. To emphasize their close relationship to tris(1-aziridyl)phosphine oxide (Tepa), these compounds are referred to as bis(1-aziridinyl)methoxy phosphine oxide to bis(1-aziridinyl)butoxy phosphine oxides, with the general structure: R—PO(N◁)$_2$; where R = OCH_3, OC_2H_5, OC_3H_7, iso- OC_3H_7 and OC_4H_9.

The compounds in the second category (phosphonic amides) had a general structure: $RNHPO(AZ)_2$; where R = methyl, ethyl, propyl, isopropyl, butyl and octyl.

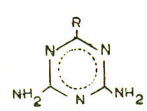
The compounds in the third category (methylmelamines) had a general structure:



where

R	R′	R″
NH_2	NH_2	$N(CH_3)_2$
NH_2	NH_2	$NHCH_3$
NH_2	$N(CH_3)_2$	$N(CH_3)_2$
$NHCH_3$	$NHCH_3$	$NHCH_3$
$NHCH_3$	$N(CH_3)_2$	$N(CH_3)_2$
$NHCH_3$	$NHCH_3$	$N(CH_3)_2$
NH_2	$NHCH_3$	$N(CH_3)_2$

The compounds in the fourth category had a general structure:



where R=methyl, chloro, isopropyl, furyl, thioyl, morpholine and pyrrolidino.

Method of Treatment. All compounds were tested as additives in the diet of newly eclosed houseflies by certain modifications in the method of Fye *et al.*[5,6] The adult flies were given candidate chemosterilants mixed in glucose for three days. For oviposition no particular medium has been used and the degree of oviposition was evaluated visually. The larvae were reared in moist bran for determining per cent hatchability at 85±3 and 80±15% R.H. in a colony room. Per cent hatchability and pupation were calculated by taking a sample of 100 eggs from each experiment. The per cent hatch of eggs was corrected by the formula of Crystal.[14]

$$\text{Corrected \% hatch} = \frac{\text{Test \% hatch}}{\text{Control \% hatch}} \times 100$$

Per cent sterility is based upon the number of pupae derived from 100 eggs. In order to find out the sex susceptibility of these compounds 50 pairs of houseflies were crossed (T♂×U♀, T♀×U♂ and T♂×T♀) in separate cages and per cent hatch and pupation were calculated as described above.

Results

Table 1 shows the effects of 5 substituted bis(1-aziridinyl)phosphine oxides and 7 substituted melamines and their results have been compared with Tepa, Hempa and Hemel. Toxicological properties of these compounds in their series could not be correlated with the sterilizing effect.

Phosphine oxides could not be tested at 1% concentration being toxic at this dose. Butoxy compound was less toxic as compared to other phosphine oxides while Hempa and Hemel proved to be much less toxic than Tepa. Oviposition was inhibited at 0.5% concentration of phosphine oxides and at 0.25% concentration the fecundity was appreciably low. Per cent sterility induced by phosphine oxides at 0.25% concentration ranged between 58–100.

Phosphonic amides (Table 3) were much active as compared to phosphine oxides.

Substituted methylmelamines were much less toxic and less effective as compared to phosphine oxides. The per cent sterility induced by them ranged between 41.1–100% at 1% concentration. The compounds N^2,N_4,N^6-trimethyl, tetramethyl and N^2,N^2,N^4-trimethyl melamine were comparatively more effective in this series. Methylmelamine showed minor effect on hatchability of eggs as compared with other methylmelamines (Table 1).

Diaminotriazines (Table 4) proved active chemosterilants of houseflies. They did not interfere much with ovipositon and with the hatchability of eggs at sterilizing doses, but the percentage sterility by these compounds was more than methylmelamines.

The 2-furyl compound was one of the most active chemosterilant in this series (Table 4).

The activity of phosphonic amides seemed to increase with the number of carbon atoms in the alkyl chain and reached a maximum when R= $CH(CH_3)_2$ but, thereafter, decreased (Table 3).

Substituted phospine oxides and phosphonic amides indicated no sex specificity and their accumulated effect on mixed sexes was greater than when sexes were treated separately (Tables 2 and 5). Substituted methylmelamines seemed to effect male flies slightly more than female (Table 2). Diaminotriazines on the other hand proved more effective in females (Table 5).

Discussion

The data on the toxicity induced by these compounds in four categories show that it is unrelated to sterilizing effect. Chang *et al.*[8] in their study concluded that the alkylating potential of a chemosterilant is alway objectionable from a toxicological point of view. The toxicity induced by phosphine oxides and phosphonic amides is much higher than methylmelamines (Table 1). This is probably due to the fact that phosphine oxides and phosphonic amides are having close relationship to tris(1-aziridinyl)phosphine oxide (Tepa) which is a very active chemosterilant.

Substituted methylmelamines being closely related to Hemel are much less toxic as well as less effective chemosterilant. DeMilo and Borkovec[15] reported Hemel less effective than some of the methylmelamines.

Among the substituted phosphine oxides the sterility induced by methoxy, ethoxy and isopropoxy compounds at 0.25% concentration was 100, 98.5 and 82.2% respectively. The activity of isopropoxy compound was more than propoxy compound. Chang and Borkovec[7] reported that in the series of bis(1-azridinyl)alkyamino phosphine oxides the sterilizing effect of the compounds decreased with the increasing length of the carbon chain in the alkyl group, except for the isopropoxy compound which was reported to be more effective chemosterilant than the ethyl compound. Chang *et al.*[8] also reported in a homologous series of phosphine oxides, the activity decreasing with increasing chain of alkyl substituent. Among phosphonic amides the activity of the compounds in the series increased with the number of the alkyl chain and it reached a maximum when R=$CH(CH_3)_2$ but thereafter it decreased. This observation is also in line with Borkovec *et al.*[12]

The sterilizing action of methylmelamines also varied much. The most effective were trimethylmelamine and tetramethylmelamine among this series, while methylmelamine proved comparatively less effective. Borkovec *et al.*[12] reported monomethylmelamine having minor effect on the hatchability of eggs in houseflies. Per cent sterility induced by methylmelamines in P.C.S.I.R.-strain ranged between 41.1–100 at 1% concentration when mixed in glucose. LaBrecque *et al.*[9] reported 100% sterility in case of methylmelamines at 0.05–1% in fly food and no hatching was noticed at 0.25–1% concentration in granulated sugar. This clearly indicates the activity of the chemosterilant changing with the diet used. They used the water-soluble salts of these compounds for 7 days while the present authors used the suspension of these compounds against P.C.S.I.R.-strain. Borkovec[2] also mentioned that

Table 1. Mortality and Sterility of Laboratory-Reared Housefly *Musca domestica*(L.), Treated Orally with Substituted Bis(1-Aziridinyl)phosphine Oxides and Melamines for Three Days Immediately After Emergence.

Compound: Ent. no.	Compound: Concn (%)	No. of flies treated	Mortality (%)	Fecundity	Corrected hatch (%)	Pupation (%)	Sterility (%)*
Substituted bis(1-aziridinylphosphine oxides)							
61342 (Methoxy)	1.0	100	100	—	—	—	—
	0.5	100	79.4	O	—	—	—
	0.25	100	29.6	L	48	0.0	100
50761 (Ethoxy)	1.0	100	100	—	—	—	—
	0.5	100	74.3	O	—	—	—
	0.25	100	35.1	L	50.2	1.5	98.5
61343 (Propoxy)	1.0	100	100	—	—	—	—
	0.5	100	54.7	O	—	—	—
	0.25	100	32.8	L	89.1	43.9	56.1
61355 (Iso propoxy)	1.0	100	91.8	O	—	—	—
	0.5	100	50.9	O	—	—	—
	0.25	100	31.8	L	38.8	17.7	82.3
61354 (Butoxy)	1.0	100	49.2	O	—	—	—
	0.5	100	14.9	L	10.8	0.0	100.0
	0.25	100	12.2	L	90.2	46.9	53.1
24915 (Tepa)	0.05	100	10.0	L	—	—	—
	0.025	100	—	L	1.91	0	100
	0.005	100	—	L	—	3.5	96.5
50882 (Hempa)	0.5	100	11.0	L	0	—	—
	0.25	100	—	L	5.7	0	100
	0.1	100	—	L	—	10.6	89.4
Methylmelamines							
22312 N^2,N^2-(Dimethyl)	1.0	100	5.4	N	60.9	8.0	92.0
50993 N^2-(Methyl)	1.0	100	0.4	N	99.1	58.8	41.2
51000 (N^2,N^2,N^4,N^4-Tetramethyl)	1.0	100	9.2	N	78.9	13.8	86.2
51035 (N^2,N^4,N^6-Trimethyl)	1.0	100	4.9	N	44.9	00.0	100.0
51239 (Pentamethyl)	1.0	100	4.3	N	49.6	10.0	90.0
51243 (N^2,N^2,N^4,N^6-Tetramethyl)	1.0	100	5.1	N	34.0	1.0	99.0
60020 (N^2,N^2,N^4-Trimethyl)	1.0	100	3.2	N	37.5	3.3	96.71
50852 (Hemel)	0.5	100	8	L	6.5	0	100
	0.25	—	—	L	—	57.0	43.0
Control	—	—	—	N	87.5	77.71	—

*Sterility is based upon the number of pupae derived from eggs.

TABLE 2. SEX SPECIFICITY OF SUBSTITUTED BIS(1-AZIRIDINYL)PHOSPHINE OXIDES AND MELAMINES AGAINST *Musca domestica* (L.), TREATED ORALLY FOR THREE DAYS IMMEDIATELY AFTER EMERGENCE (AVERAGE OF THREE REPLICATES).

Compound		Sex treated	Fecundity	Corrected hatch(%)	Pupation (%)	Sterility (%)*
Ent. No.	Concn(%)					
Substituted bis(1-aziridinyl)phosphine oxides						
61342	0.25	M	L	92.5	47.3	52.7
(Methoxy)	0.25	F	L	83.7	51.5	48.5
	0.25	Both	L	44.7	00.0	100.0
50761	0.25	M	L	53.8	15.6	84.4
(Ethoxy)	0.25	F	L	45.5	6.4	93.6
	0.25	Both	L	43.8	1.5	98.5
61343	0.25	M	N	94.1	61.2	38.8
(Propoxy)	0.25	F	N	90.3	62.1	37.9
	0.25	Both	L	82.9	43.9	56.1
61355	0.25	M	L	36.6	19.7	80.3
(Isopropoxy)	0.25	F	L	38.8	24.3	75.7
	0.25	Both	L	36.1	17.7	82.3
61354	0.25	M	N	89.6	76.0	24.0
(Butoxy)	0.25	F	N	86.0	53.1	46.9
	0.25	Both	L	84.0	46.9	53.1
24915	0.005	M	L	—	2.2	97.8
(Tepa)	0.005	F	L	—	17.5	82.5
	0.005	Both	L	—	3.5	96.5
50882	0.1	M	N	—	11.0	89.0
(Hempa)	0.1	F	N	—	74.2	25.8
	0.1	Both	L	—	10.6	89.4
Methyl Melamines						
22312	1	M	N	68.3	19.6	80.4
(N^2,N^2-Dimethyl)	1	F	N	75.5	22.6	77.4
	1	Both	N	56.6	8.0	92.0
50993	1	M	N	99.8	65.6	34.4
(N^2-Methyl)	1	F	N	89.7	72.3	27.7
	1	Both	N	90.4	58.8	41.2
51000	1	M	N	95.2	38.3	61.7
(N^2,N^2,N^4,N^4-Tetramethyl)	1	F	N	97.1	31.8	68.2
	1	Both	N	84.1	13.8	86.2
51035	1	M	N	57.7	17.6	82.4
(N^2,N^4,N^6-Trimethyl)	1	F	N	81.4	31.0	69.0
	1	Both	N	41.8	00.0	100.0
51239	1	M	N	60.7	20.5	79.5
(Pentamethyl)	1	F	N	75.1	21.1	78.9
	1	Both	N	45.9	10.0	90.0
51243	1	M	N	46.1	13.1	86.9
(N^2,N^2,N^4,N^6-Tetramethyl)	1	F	N	65.1	19.3	80.7
	1	Both	N	31.6	1.0	99.0
60020	1	M	N	69.1	12.6	87.4
(N^2,N^2,N^4-Trimethyl)	1	F	N	68.9	13.9	86.1
	1	Both	N	34.6	3.3	96.7
50852	0.25	M	N	—	72.3	27.7
(Hemel)	0.25	F	N	—	72.5	27.5
	0.25	Both	L	—	57.0	43.0
Control	Both Untreated	N	94.0	83.1	—	

*Sterility is based upon the number of pupae derived from eggs. L, low; N, normal.

TABLE 3. STERILIZING EFFECTS OF PHOSPHONIC AMIDES IN HOUSEFLY *Musca domestica* (L.) AFTER 3 DAYS ORAL TREATMENT IMMEDIATELY AFTER EMERGENCE.

Compounds: Ent. no	Compounds: Concn %	Corrected mortality (%) (after 7 days)	Fecundity	Corrected hatch (%)*	Pupation (%)*
51254	0·5	88			
(Methyl)	0·25	43·5	None		
	0·1	29·5	None		
	0·01	5·3	L	0(0)(0)	0(0)(0)
	0·005	3·5	L	0(0)(0)	0(0)(0)
	0·001	2·0	L,M	66.5, 86.5, 87.5	40, 59, 71
50787	0·5	91·5			
(Ethyl)	0·25	29	None		
	0·1	9	L	0(0)(0)	0(0)(0)
	0·01	11·2	L	0(0)(7)	0(0)(3)
	0·005	8	L,M,N	0(0)(0)	0(0)(0)
	0·001	1	L,M,N	66.5, 80.5, 89.5	35,52.5,7
51253	0·5	100			
(Propyl)	0·25	100			
	0·1	100			
	0·01	48	None		
	0·005	13·9	L	0(0)(6)	0(0)(4)
	0·0025	8·5	L,M,N	28,(50)73	10, 18.5, 34
51256	0·5	81·5			
(Isopropyl)	0·25	42·5	None		
	0·1	17·0	L	0(0)(0)	0(0)(0)
	0·01	9·6	L,L,M	0(4)(6)	0(2)(3)
	0·005	5·0	L,M,N	0(0)(0)	0(0)(0)
	0·001	2·0	L,M,N	82.5,87.5,91	56.5,60.5,78
51028	0·5	38	None		
(Butyl)	0·25	16	L	0(0)(0)	0(0)(0)
	0·1	14·5	L	0(0)(15)	0(0)(9)
	0·05	5·9	L,M,N	0(5)(10)	0(2)(5)
	0·01	2·0	M,N,N	32, 50, 76	9, 23, 49
50788 (Octyl)	0·5	4·2	M,N,N	72, 82, 86	14, 44, 55
Control		0	N	98,99,98	96,99,96

TABLE 4. EFFECTIVENESS OF DIAMINOTRIAZINES ON THE EGG PRODUCTION, HATCHABILITY AND PUPATION IN HOUSEFLY *Musa domestica* (L.) AFTER 3 DAYS ORAL TREATMENT IMMEDIATELY AFTER EMERGENCE.

Compound: Ent. no.	Compound: Concn (%)	Corrected mortality (%) (after 7 days)	Fecundity	Corrected hatch (%)*	Pupation (%)*
50715 (Methyl)	0·5	3·2	N	90,85,94	75,83,39
50982 (Chloro)	0·5	2·1	N	97,92,98	90,90,95
22657	0·5	3·2	N	90,90,89	0,0,0
(Isopropyl)	0·25	2·5	N	81,82,92	48,58,83
22641	0·5	54	None		
(Furyl)	0·25	11	L	0,85,90	0,0,0
	0·1	4	L	89,88,91	0,0,0
	0·05	2	L,M,N	52,89,90	0,0,0
	0·01	0	L,M,N	85,98,95	0,10,51
60103	0·5	19	L	98,97,96	0,0,0
(Thionyl)	0·25	8	L,M,N	92,93,95	0,0,0
	0·1	4·5	L,M,N	92,80,97	0,0,0
	0·05	3	L,M,N	94,95,96	0,40,55
51014	0·5	16	L	98,98,98	0,0,4
(Morpholino)	0·25	5·9	N	95,87,90	0,0,0
	0·1	2	N	73,80,89	0,15,38
	0·05	0	N	98,96,95	26,57,89
51112	0·5	12·5	L	97,97,95	0,0,9
(Pyrrolidino)	0·25	8.5	L	95,98,98	0,0,0
	0·1	5·5	L,M,N	81,85,90	0,5,12
	0·05	0	L,M,N	80,90,95	22,48,76
	0·01	0	M,N	95,90,94	39,54,68
Control		0	N	98,99,98	96,99,6

TABLE 5. SEX SPECIFICITY OF PHOSPHONIC AMIDES AND DIAMINOTRIAZINES IN *Musca domestica* (L.)

Compound: Ent. no.	Compound: Concn (%)	Sex trated	Fecundity	Corrected hatch (%)*	Pupation (%)*
51254	0·001	♂	L,M,N	87,91,98	49,60,94
		♀	L,M,N	70,92,97	53,42,82
50787	0·001	♂	L,M,N	77,92,94	43,66,73
		♀	L,M,N	91,92,98	68,75,81
51253	0·0025	♂	M,M,N	36,57,87	13,19,34
		♀	M,M,N	75,70,92	31,39,48
51256	0·001	♂	M,M,N	90,98,98	80,84,98
		♀	M,M,N	92,90,95	67,61,91
51028	0·01	♂	M,M,N	53,69,89	14,27,56
		♀	M,M,N	61,78,91	21,39,85
50788	0·5	♂	N	92,81,93	37,66,82
		♀	N	83,91,96	42,65,75

Compound: Ent. no.	Compound: Concn (%)	Sex trated	Fecundity	Corrected hatch (%)*	Pupation (%)*
Diaminotriazines					
50715	0·5	♂	N	90,91,95	81,85,92
		♀	N	89,90,95	78,83,92
50982	0·5	♂	N	96,96,98	90,95,95
		♀	N	98,97,98	96,95,96
22657	0·25	♂	N	80,84,98	57,68,95
		♀	N	98,96,96	35,52,90
22641	0·01	♂	L,M,N	94,82,98	53,54,39
		♀	L,M,N	50,89,97	0,22,65
60103	0·05	♂	N	88,98,98	48,83,91
		♀	N	98,95,93	10,41,89
51014	0·05	♂	N	94,98,84	91,96,65
		♀	N	98,98,95	91,93,78
51112	0·01	♂	L,M,N	97,98,96	85,93,90
		♀	L,M,N	85,93,91	57,82,78
Control	—	—	N	98,99,98	96,99,96

*Results of triplicated experiments. L, low, M, moderate.

the poorly soluble or insoluble chemosterilants are much more limited in their action. According to Borkovec and DeMilo[11] the biological activity is known to vary widely due to striking differences between solubilities of the basic melamines. They tested such compounds in the form of hydrochlorides and reported that their activity increased with more uniformity.

Chang and Borkovec[8] observed that the sterilizing activity of melamines increase with the degree of methylation, but the maximum activity is reached with tetramethylmelamine rather than with hexamethylmelamine (Hemel). In the present study the activity of Hemel was more than that of all the methylmelamines. This difference again may be due to the insoluble melamines used here.

Diaminotriazine did not interfere with oviposition and with the hatchability of eggs (Table 4), but the per cent sterility by these compounds was more than methylmelamines. According to Borkovec *et al.*[12] the physical and chemical properties of melamines are similar, although diaminotriazines are weaker bases and are effective primarily in the females without interfering with oviposition and with the hatchability of eggs.

Among the diaminotriazines, the compound (25) 2,4-diamino-6-(2-furyl)-*s*-triazine proved most effective in this series. Borkovec *et al.*[12] in their study also referred to this compound as extremely active in houseflies while this very compound was observed to be inactive in screwworm, the Mexican fruitfly and boll weevil.

The data on sex specificity indicate that phosphine oxides and phosphonic amides effected both the sexes more or less equally, but their accumulated effect on mixed sexes was greater than when sexes were treated separately (Tables 2 and 5). According to Borkovec and Woods[16] the derivatives of aziridines are known for their male sterilizing activity which is often coupled with female-sterilizing activity.

The results with substituted methylmelamines indicated that they sterilized male flies slightly more than females (Table 2). Borkovec and DeMilo[11] also observed that as a group only the methylated melamine sterilized male flies, while the melamines with cyclic substituents could not sterilize males. In the present study the effect of Hemel was more on male flies. This observation is in line with Borkovec[17] and Fye *et al.*[6]

Acknowledgements. The authors are indebted to the kindness of Dr. A.B. Borkovec, Incharge, Chemosterilant Investigations, U.S.D.A., Beltsville, Md., U.S.A., for the generous supply of chemicals and for his valuable suggestions.

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