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Investigation into the Problem of Insecticide Sorption by Soils

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In view of the importance of the problem of insecticide sorption by certain soils, and its possible implications for mosquito control, an investigation was carried out on the influence of some factors governing sorption.

In experiments with thin layers of homogenized soil/insecticide mixtures, in which no diffusion took place, changes in relative humidity had a pronounced influence on the effectiveness of the insecticide, a 10% increase in humidity doubling the toxicity. Further experiments using DDT labelled with ^{14}C *showed that the effect of humidity was the only factor influencing toxicity.*

Studies with ^{14}C *DDT and bio-assay determination of non-labelled DDT and dieldrin showed that movement of the insecticide in the soil was blocked at both very high and very low humidity, and that inward migration was found only at intermediate humidities. It was also observed that the migration of water in the soil caused the insecticide to move in the same direction.*

As at a high relative humidity the inward migration of the insecticide is blocked, and as the initial loss in effectiveness by sorption is counterbalanced by the greater availability of the remaining toxicant, the application of the usual field dosage of dieldrin (0.5 g/m²) remained effective for a considerable period. This would suggest that sorption might well be a problem in the field only when humidity is low. In work on means of reducing sorption under dry conditions, encouraging results were obtained with wettable powders based on ground solidified melts of dieldrin and certain resins.

INTRODUCTION

Under the aegis of the World Health Organization large amounts of insecticides are regularly used in malaria eradication campaigns. In tropical regions where malaria is endemic, the native " mud-huts ", mainly constructed of local soils, are sprayed internally for this purpose, but the residual effectiveness obtained has been, for reasons unknown, not always as good as anticipated.

Hadaway & Barlow (1951) and simultaneously Downs, Bordas & Navarro (1951) found in the laboratory that solid particles of DDT disappeared from the surface of various types of soils, such as are used in the construction of mud huts, by a process of physical sorption. This process seems to occur with all commonly used residual insecticides, the rate of sorption being higher with the more volatile chemicals (Hadaway & Barlow, 1952). This results in a great decrease in effectiveness, which is, however, less under humid than under dry conditions (Barlow & Hadaway, 1956, 1958a, 1958b). The sorption of insecticides into soils might also cause a considerable loss of residual effectiveness under field conditions (Bordas & Navarro;[2] Burnett, 1956, 1957; Langbridge;[3] Pal & Sharma, 1952).

The investigation reported in this paper is a study of the influence of certain factors on the process of sorption, and particularly of the influence of relative humidity on the distribution and availability of the insecticide in the soil, and of the influence of formulation on the persistence of the insecticide on the soil surface.

No attempts have been made to correlate the sorption of insecticides with the physical properties

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[1] Shell Research Ltd., Woodstock Agricultural Research Centre, Sittingbourne, Kent, England.

[2] *Studies on the vapour toxicity, repellency and residual activity of DDT, chlordane, lindane and dieldrin* (unpublished working document WHO/Mal/125; WHO/Insecticides/38).

[3] In: Nigeria, Federal Malaria Service, *Western Sokoto Malaria Control Pilot Project, Insecticide Chemistry Laboratory, Annual report 1955-56*, Yaba-Lagos (mimeographed document).

(Bertagna, 1959) of the substrate. Sorptivity was merely determined by applying the insecticide and measuring the de-activation of the insecticidal deposit by bio-assay.

MATERIALS AND METHODS

Preparation of soil panels

Although the soils are often mixed with materials such as sand, straw and cow dung in the construction of mud huts, it was thought preferable to use pure soils in these experiments. Test panels were prepared by mixing the soil with water and moulding the paste in a metal ring (1 cm high, 10 cm diameter). In most of the experiments a red laterite soil from Tanganyika (Babati) was the standard material. After drying and storage at a temperature of 30°C for several weeks, the panels were sprayed by means of a De Vilbiss hand sprayer. Unless mentioned otherwise, the insecticide was applied as a 1% suspension in water, at a dosage of 3 mg active material per dm^2 (25 mg per square foot). The panels were stored at a temperature of 30°C (± 2°C) and a relative humidity of under 40%.

At regular intervals the residual activity was tested by exposure of insects, employing three different techniques.

Short-exposure technique

This technique was based on the exposure of houseflies (*Musca domestica*). These insects are very active and will pick up the insecticide in comparatively large quantities in a very short time; they are therefore extremely useful in the comparison of insecticidal surface deposits. The procedure was as follows. From the stock of flies, 30-40 were allowed to escape into a test tube, which was then placed over a small opening in a thin stainless steel plate covering an empty Petri dish (diameter 9 cm). After transfer of the flies the Petri dish was placed, metal plate downwards, on the surface of the test panel and the cover plate withdrawn for 30 seconds. Then the plate was re-inserted, and the flies were anaesthetized by introducing carbon dioxide through the hole in the plate, and transferred to 250-ml observation bottles, containing some sugar and covered with muslin. Knockdown readings were taken at hourly intervals up to 6 hours after exposure, and after 24 hours the total number of moribund and dead flies was recorded as mortality. Control mortality was always lower than 4% and therefore not taken into account. Tests were done at room temperature.

Long-exposure technique

For the exposure of mosquitos (*Aëdes aegypti*) a similar technique to that used by Hadaway & Barlow was adopted. Thirty mosquitos (sex ratio 1:1, if not otherwise specified), fed with sugar water only, were exposed to soil panels under glass funnels for various times. As the mosquitos are unable to rest on clean glass at the reverse slope under the funnels, this is virtually a " no-choice " exposure method. After exposure for various times up to 4 hours, the mosquitos were transferred to recovery bottles, in which a piece of paper provided resting sites. A plug of cotton wool soaked in sugar solution was inserted in a central aperture in the covering muslin. Mortality was recorded after 24 hours. Tests were made at 23°-24°C.

Tests in exposure chambers

When exposing mosquitos to the horizontal surfaces of the soil panels, it was noticed that the insects were very active and walked on the panel nearly continuously throughout the exposure period. This is quite different from their normal behaviour. When free, they move about almost entirely by flight, make contact with the surface only by means of a number of distinct landings, and hardly walk at all. This behaviour could be secured by using an exposure technique similar to that developed in the Technical Development Laboratories of the US Public Health Service at Savannah, Ga. Use is made of relatively large plywood boxes of which only the side panels (3 inches × 12 inches, or 7 cm × 30 cm) are plastered with a layer of soil and subsequently sprayed with insecticide. As the top and bottom parts are left untreated, this method is a " free-choice " exposure method. The insect population density during exposure is considerably less than in the other techniques; moreover, the insects are exposed to the surface in a vertical position, and in the dark. The holding of mosquitos after exposure was similar to that in the " long-exposure " technique.

Insecticides

Some of the experiments were carried out with non-formulated insecticides: dieldrin (90% HEOD + 10% related compounds), aldrin (95% HHDN), DDT (100% *p,p'*-isomer), BHC (100% γ-isomer). The dieldrin and BHC wettable powders used in some of the experiments contained 50% active material, and were prepared at the Woodstock Agricultural Research Centre. The DDT 50% wettable powder was a " Murphy " product. The

TABLE 1

TOXICITY TO *MUSCA DOMESTICA* OF LOW DOSAGES OF DIELDRIN WETTABLE POWDER ON BABATI SOIL

Dosage (mg/dm^2)	Percentage knockdown after hours shown						
	1½	2	3	4	5	6	24
3	29	58	100				100
1	22	35	65	100			100
0.3		0	6	23	46	63	100
0.1		0	3	13	22	34	94
0.03					0	7	40
0.01						0	14

^{14}C DDT used had a melting point of 104°-105°C, specific activity 0.5 mc/g.

SURFACE LIFE OF THE INSECTICIDE IN THE PARTICULATE STATE

As can be observed visually, insecticide particles may disappear from the surface of dry soils within a few days. For the estimation of this period, quantitative chemical analytical methods were considered too elaborate, and visual assessment not sufficiently accurate on account of the difficulty of distinguishing the insecticide from the minerals of the substrate and from the filler in the case of wettable powders. It was therefore decided to employ a bioassay technique for the evaluation of persistence.

Table 1 gives the results of 30 seconds' exposure tests with houseflies, using very low dosages of dieldrin 50% wettable powder on panels of Babati soil, tested immediately after application of the insecticide. Dieldrin deposits as low as 0.1 mg/dm^2, which is about 3% of the standard dosage (3 mg/dm^2), still gave a 24-hour mortality of 94%. Therefore a fall in mortality below 90% was accepted as an indication that the particulate insecticide had virtually disappeared. This criterion of the end of "surface life" can, strictly speaking, be used only with insecticides of the same order of toxicity.

Short-exposure tests on soil panels, using the standard dosage of dieldrin 50% wettable powder, showed that the sorption rate is very high (Table 2). On Babati soil particulate dieldrin did not persist for longer than one day. On Mexico, D.F., soil, which is a markedly less sorptive material, the persistence was about 3 days.

TABLE 2

DIELDRIN SORPTION BY VARIOUS MATERIALS, EXPRESSED AS PERCENTAGE 24-HOUR MORTALITY AFTER 30 SECONDS' EXPOSURE OF *MUSCA DOMESTICA*

Substrate	Age of residue					
	2 hours	1 day	2 days	3 days	1 week	2 weeks
Wood	100			100	100	100
Paper	100			100	100	32
Brick	100			48	50	76
Plaster	100			57	38	
Mexico, D.F., soil	100	100	96	98	82	35
Babati soil	100	96	55	44	29	0

Other building materials, such as wood, bamboo and leaves, have been reported to be comparatively non-sorptive (Barlow & Hadaway, 1957; [1] Hadaway & Barlow, 1956, 1958 [2]). Test materials included brick, plaster, paper and wood, which may be used in both tropical and temperate areas. Except for wood, these materials all sorb particulate dieldrin rapidly, brick and plaster appearing to be as sorptive as Babati soil, while paper was somewhat less active, permitting a surface life of particulate dieldrin of the order of one to two weeks.

Sorption of solid insecticides is thus not restricted to soils, but can be regarded as a general phenomenon.

The rate at which a given particle of insecticide is sorbed can be calculated. This has been done by Barlow & Hadaway (1955), who found a reasonably good agreement between the calculated and the visually observed life of DDT particles.

In the present work the residual life of dieldrin, aldrin, γ-BHC and DDT was estimated by bioassay and compared with the calculated life. Use was made of insecticide suspensions of known particle size, sprayed on Babati soil panels at a dosage of 3 mg active material per dm^2. Calculation was made on the assumption that disintegration takes place predominantly from the side of the particle which is

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[1] Unpublished document CIRU/Porton/Report No. 139 (Colonial Insecticides Research Unit, Porton Down, Wilts, England).

[2] Unpublished document CIRU/Porton/Report No. 155 (Colonial Insecticides Research Unit, Porton Down, Wilts, England).

TABLE 3

OBSERVED AND CALCULATED PERSISTENCE OF INSECTICIDES ON BABATI SOIL AT 30°C

Insecticide	Volatilization rate [a] under high vacuum (g/cm ^{2}/sec)	Approximate particle size (μ)	Persistence	
			Found by bio-assay	Calculated
DDT	3.3×10^{-9}	10	ca. 3 days	5.8 days
Dieldrin	1.5×10^{-8}	10	1-1½ days	1.3 days
Dieldrin	1.5×10^{-8}	50	ca. 1 week	6.5 days
Aldrin	4.7×10^{-7}	10	ca. 1½ hours	0.9 hour
γ-BHC	3.8×10^{-7}	50	6-8 hours	5.9 hours

[a] Measured by Langmuir's effusion method.

in actual contact with the sorbent and that the conditions for sorption are similar to those of evaporation under high vacuum. The results are listed in Table 3, and it can be seen from the figures that there was a fairly good agreement between observed and calculated values.

The above results confirm that the rate of volatilization and particle size are correlated with the persistence of the insecticidal particle. This is in line with the results obtained by Barlow & Hadaway.

In the usual wettable powders the majority of the particles are below 10 μ, and consequently, the surface life of the particulate insecticide on a sorptive surface is short. Effectiveness in the field, however, is maintained for a very long time (several months), and it must therefore be assumed that under the prevailing conditions toxicity is caused by the insecticide in a non-particulate state.

EFFECTIVENESS OF THE INSECTICIDE IN THE SORBED STATE

It has been found in earlier investigations (Gerolt, 1958) that dieldrin, BHC and DDT do not act specifically in the vapour phase. It is therefore assumed that the insecticide, after being absorbed by the soil and subsequently adsorbed on the surface of the soil particles (internal surface), can be taken up by the insects by direct contract.

As the insecticide diffuses deeper into the soil the actual concentration at the internal surface in the top layer rapidly decreases to a low level. For example, penetration of dieldrin from an initial deposit of 3 mg/dm^2 to a depth of 0.1 mm in soil (corresponding to 2 g of soil/dm^2), the internal surface area of the soil being 40 m^2/g soil, will lead to an average concentration at the internal surface of 0.4 μg/dm^2. It is therefore not surprising that long exposure is required to show a toxic effect. Long contact periods are, however, common in the field.

An experiment was carried out with the aim of obtaining information on the residual effectiveness of dieldrin in the sorbed state, using a selection of different soils as substrate. The soil panels were sprayed with a dieldrin 50% wettable powder at a dosage of 3 mg dieldrin/dm^2, the residual effect of which was tested at intervals. The panels were stored at a constant relative humidity of 50%. Adult *Aëdes aegypti* mosquitos (30 per exposure; sex ratio 1 : 1) were used in the exposure tests, using the "long-exposure" technique. The results are shown in Table 4, and it can be seen that the effectiveness of dieldrin in the sorbed state may last for a considerable period under conditions where insects are subject to long exposure. The rate of de-activation varied according to the soil used, and the standard Babati soil appeared to be of medium activity in this respect. De-activation was, however, virtually complete within 15 weeks for all soils, with the possible exception of Mexico, D.F., soil.

INFLUENCE OF HUMIDITY

There is ample evidence from both laboratory and field experiments that the effectiveness of insecticides on sorptive soil surfaces varies considerably with the relative humidity of the atmosphere, and there are indications that the influence of humidity can be at least threefold:

(*a*) influence on the initial sorption of the particulate insecticide;

(*b*) influence on the rate of inward diffusion;

(*c*) influence on the availability of the sorbed toxicant to the insect.

Initial sorption of particulate insecticide

As observed visually by Barlow & Hadaway (1958b), the rate of initial sorption of dieldrin from the particulate state decreases with increasing humidity, particularly on soils of rather low sorptive capacity. They found that dieldrin particles (10 μ or less; dosage 11 mg dieldrin/dm^2) on highly sorptive soil disappeared in less than one day at low relative humidity (10%) and in a few days at high humidity (90% RH). On less sorptive soils this difference in persistence is considerably greater—a few days and over seven weeks respectively.

TABLE 4

DE-ACTIVATION OF DEPOSITS OF DIELDRIN WETTABLE POWDER OF VARIOUS SOILS, EXPRESSED AS PERCENTAGE 24-HOUR MORTALITY OF *AËDES AEGYPTI*

Time after spraying	3 days		1 week		2 weeks	10 weeks		15 weeks
Exposure time Origin of soil	4 min.	16 min.	16 min.	1 hour	1 hour	1 hour	4 hours	4 hours
Tanganyika (Babati)	86	100		86	82	0	100	10
Uganda (Entebbe)	14	33	2	38	7	0	83	13
Nigeria (Lagos)	95	100	3	67	50	0	100	14
USA (Colorado)	100	100	100	100	100	0	100	0
USA (Savannah, Ga.)	14	56	2	34	11	0	17	10
Mexico (Michoacán)	85	95	27	60	35	3	97	18
Mexico (Morelos)	100	100	100	100	93	3	100	28
Mexico (Mexico, D.F.)	100		90	100	80	42	100	44

The influence of humidity on the rate of initial sorption was confirmed in our experiments by means of bio-assay. The results indicated a surface life of dieldrin deposits (9 mg dieldrin/dm^2) on Babati soil of the order of 3 days at 10% RH and about 3 weeks at 90% RH.

Rate of inward diffusion

As regards the influence of humidity on further migration, the same authors (Barlow & Hadaway, 1955, 1958b) found by chemical analyses of scrapings of treated Uganda soil panels that the inward diffusion of the insecticide in the soil after the initial sorption is more rapid at higher relative humidity. Miles & Pearce (1957), however, in their experiments with ^{14}C DDT on Savannah river mud, found the opposite.

In view of this controversy it was considered advisable to carry out some further work on this subject. Since no appreciable effect of humidity upon the action of the insecticide in the insect is to be expected (Hadaway & Barlow, 1957), it was assumed that differences in the toxic effect, when measured under the same conditions (i.e., the same soil, relative humidity and temperature), are caused by differences in concentration.

The experiment included two pairs of replicate soil panels, sprayed with dieldrin 50% wettable powder at the standard rate of 3 mg dieldrin/dm^2, which were stored at 10% and 90% relative humidity respectively for a period of over half a year. Those originally stored at 10% RH were first re-stored at 50%, and then, after testing, at 90%. Panels originally stored at 90% were re-stored at 50% and 10% respectively. Table 5 gives the results of the exposure tests carried out after the various periods of storage at different humidities. A comparison is given with results obtained previously with a panel continuously stored at 50% relative humidity and re-stored at 90%.

Re-storage for 2 weeks at 50% relative humidity resulted in about the same toxicity irrespective of whether the panels had been previously stored at 10% or 90% RH. This suggests that the concentration of insecticide in the top layer must have been very similar, and thus that the diffusion rate in both cases was approximately the same. In comparison, panels kept at 50% and subsequently re-stored at 90% showed the lowest toxicity figure. Moreover, this lower effectiveness was obtained in a shorter period.

These observations would indicate that the rate of inward diffusion is higher at 50% than at 10% relative humidity, thus confirming the results of Barlow & Hadaway (1958a). It is also higher than at 90%, a finding which could be linked with their results of scraping analyses indicating that at 90% relative humidity the insecticide remained in the top layer. This effect was observed with DDT on Uganda soil, but not with dieldrin or BHC. The

TABLE 5

INFLUENCE OF HUMIDITY ON THE EFFECTIVENESS OF DIELDRIN ON SOIL PANELS, EXPRESSED AS PERCENTAGE 24-HOUR MORTALITY OF *AËDES AEGYPTI* AFTER 4 HOURS' EXPOSURE

Storage conditions	Relative humidity				
	10%	50%	90%	50%	10%
25 weeks at 10% RH	0-7				
29 weeks at 10% RH +2 weeks at 50% RH		10-42			
29 weeks at 10% RH +2 weeks at 50% RH +2 weeks at 90% RH			100-100		
25 weeks at 90% RH			100-100		
29 weeks at 90% RH +2 weeks at 50% RH				7-11	
29 weeks at 90% RH +2 weeks at 50% RH +2 weeks at 10% RH					0-0
15 weeks at 50% RH		10			
17 weeks at 50% RH +1 week at 90% RH			4		

maximum rate of diffusion with DDT was observed at 80% relative humidity. We assume that this exceptional behaviour of DDT at this high humidity was caused by capillary condensation of water, taking place to such a degree that it prevented the insecticide from penetrating deeper into the soil. However, it is not clear why this should occur with dieldrin on Babati and with DDT on Uganda soil, but not with dieldrin on Uganda soil.

As can be seen from Table 5, re-storage of the 50% relative humidity panels at 90% did not increase toxicity. This has been further confirmed in an experiment in which the various soils mentioned in Table 4 had been subjected to 3 weeks' re-storage at 90% RH. Apparently, in the majority of the soils examined, the concentration of toxicant in the top layer had become so low during the prolonged storage at 50% RH and subsequent storage at high humidity that no re-activation response could be observed. A slight re-activation was noticed only in the test with Mexico, D.F., soil.

Availability of sorbed toxicant

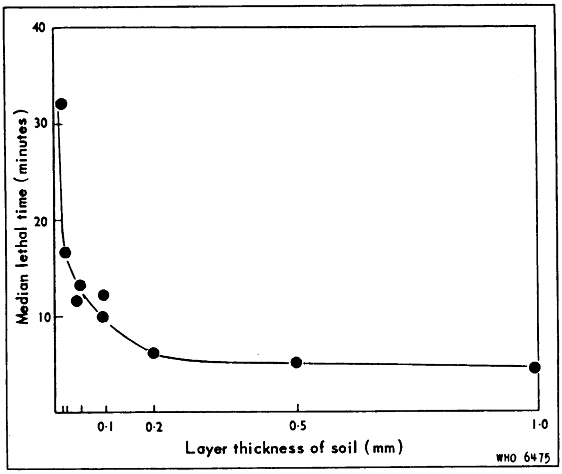
According to Barlow & Hadaway (1956), the increasing rate of inward diffusion at higher humidity is caused by an increase in mobility of the insecticide molecules on the internal soil surface owing to preferential adsorption of water. This greater mobility not only results in an increased rate of inward diffusion and a consequent decrease in concentration in the top layer, but it also makes any given quantity of toxicant more easily available to the insects. Barlow & Hadaway also showed that the effect of humidity on the availability of the insecticide, and consequently on its effectiveness, is based on a reversible process. One may therefore assume that the final toxicity is governed by two factors, concentration and availability, which are both influenced by changes in humidity; but our knowledge of the relative importance of each of these factors is still fragmentary.

An attempt has been made in this work to separate the two factors. We examined the influence of humidity on the availability of the toxicant while keeping the concentration constant, thus excluding the factor of inward diffusion. This could be realized in experiments with soil homogeneously mixed with a given quantity of dieldrin. It was assumed that after a relatively short time the dieldrin would be evenly distributed in the molecular state, adsorbed on the soil particles. The use of panels was considered a drawback in this case, as it takes a long time to achieve an equilibrium between water content of the soil and atmospheric humidity. Experiments in which the water content of successive layers of a soil panel was measured showed that after transference from 10% to 90% relative humidity, the equilibrium in the top layer was not reached within 4 weeks. To reduce the time needed to attain moisture balance, very thin layers were employed. A calculated amount of soil paste was therefore applied to a glass plate, so that after drying a soil layer of known and even thickness, firmly attached to the glass surface, was obtained.

The mobility of the insecticide on the internal surface of the soil permits replacement from deeper layers when toxicant is taken up by the insects, but to enable an unrestricted replacement a certain minimum thickness of layer is required. In order to determine this minimum, tests were carried out with soil layers of various thicknesses. Dieldrin 50% wettable powder had been mixed homogeneously with the soil beforehand at a dosage of 1 mg of dieldrin per g of dry soil. It proved necessary to

FIG. 1

EFFECT OF LAYER THICKNESS ON AVAILABILITY OF DIELDRIN IN BABATI SOIL



remove the very coarse material from the soil (over 250 μ) and the specific internal surface area was then found to be about 100 m²/g of dry soil; the final concentration was estimated to be roughly 0.1 μg dieldrin/dm². A sufficiently long period was allowed for conditioning the mixture. Tests were carried out at 89% RH.

Exposure tests with *Aëdes aegypti* (30 females per exposure) showed a clear correlation between toxicity, as expressed in median lethal exposure time (time to give 50% kill; MLT), and layer thickness (Fig. 1). It was shown that the layers must be at least 0.2 mm thick to give the maximum toxicity (minimum MLT); below that thickness, toxicity was impaired.

The influence of humidity on the availability of dieldrin was estimated in the way described above, using layers of 0.5 mm thickness. Fig. 2 shows that there was a direct correlation between log MLT and relative humidity, any reduction of humidity by 10% doubling the MLT. It is obvious that the influence of humidity on the effectiveness of dieldrin is much more pronounced than in experiments with the usual soil panels. Whereas at 90% relative humidity only 10 minutes' contact is required to cause 50% mortality, about 10 hours would be necessary to obtain the same effect at 30%.

EXPERIMENTS WITH ^{14}C DDT

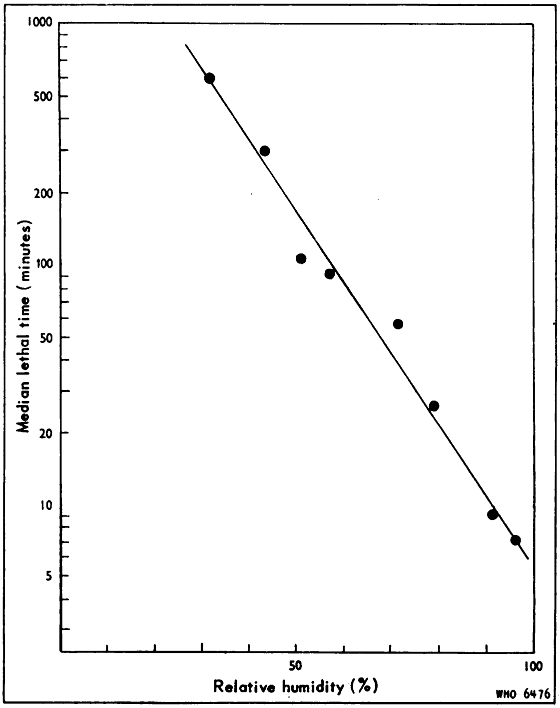
Experiments with homogenized insecticide/soil mixtures

It has been assumed in the experiments with homogenized soil/dieldrin mixtures that any increase in effectiveness at higher humidity was caused only by the greater availability of the insecticide. However, no strict proof has so far been given that absolutely no differences in concentration of the insecticide in the thin soil layer occurred. Therefore, further experiments were designed in which concentration was the only factor measured. For this purpose the use of labelled insecticide was considered essential.

The only reference to experiments of this type is from Miles & Pearce (1957). They studied the effect of sorption of crystalline deposits of ^{14}C DDT (200 mg per square foot, or 2 g per m²) on Savannah river mud panels, utilizing a gas flow counter, and found increased radioactivity after transfer from low to high humidity. They concluded that a rise in humidity causes re-migration of the insecticide towards the surface, which could suggest that the effect of improved toxicity is due to an increase in concentration of the insecticide in the top layer. This is in line neither with the theory of Barlow & Hadaway nor with our expectations.

FIG. 2

EFFECT OF RELATIVE HUMIDITY ON AVAILABILITY OF DIELDRIN IN BABATI SOIL



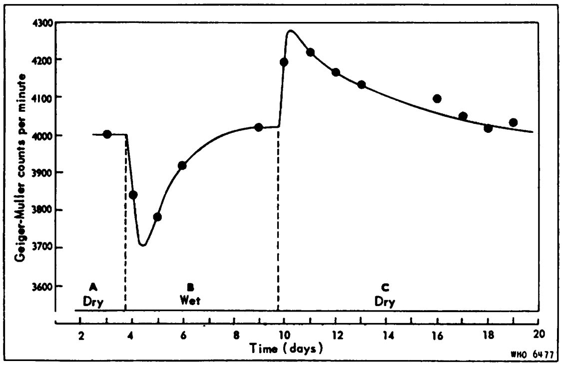
In order to obtain more information on this matter a similar experiment was carried out at the Woodstock Agricultural Research Centre, but with the insecticide evenly distributed over the entire internal surface of the soil. Use was made of ^{14}C DDT which was mixed in a hexane solution with Babati soil (particle size $<250\mu$; internal surface area about 80 m^2/g), the concentration being 8 mg of DDT per g of dry soil (concentration at the internal surface roughly 1 $\mu g/dm^2$). After evaporation of the hexane, a layer approximately 0.75 mm thick was mixed with water and applied as a paste to an aluminium planchet of 22 mm diameter. After storage in dry conditions for 3 weeks the radioactivity was measured at low and high humidity.

To avoid any interference from variations in atmospheric conditions, the source was kept continuously in the lead-chamber (it was not touched throughout the experiment), dry and humid air being supplied through a channel in the bottom plate at a constant rate of approximately 10-15 litres per hour; the temperature was kept at 23°-24°C. Counting was done with a thin end-window Geiger-Müller tube. Each measurement included at least 30 000 counts. ^{14}C is a weak β-source, the maximum range of the particles in soil being about 0.15 mm and 90% being absorbed by a soil layer of only 0.05 mm. Any noticeable change in radioactivity would therefore indicate a change in concentration of insecticide within the uppermost layer of 0.05 mm. The results (in counts per minute) are given in Fig. 3. Parts A and C indicate periods of low humidity (air dried by silica gel); B shows a period of high humidity (air almost saturated with water vapour).

Only under conditions of actual sorption or desorption of water did temporary changes in concentration of the insecticide in the top layer occur. Sorption of water resulted in a decrease in concentration (" washing in " effect), desorption in an increase (" washing out " effect). In other words, the insecticide moved with the moving water. However, under constant conditions, no matter at what level of humidity, the distribution of the insecticide was always the same. This shows with certainty that the greater toxic effect at high humidity cannot be attributed to a higher concentration at the surface, for, as is clear from Fig. 3, the concentration is never greater than the original value. The greater toxic effect must therefore be entirely due to an increased availability of insecticide to the insect. A similar reasoning applies to conditions of low humidity.

FIG. 3

INFLUENCE OF CHANGE IN RELATIVE HUMIDITY ON CONCENTRATION OF ^{14}C DDT IN TOP LAYER OF BABATI SOIL



It is also noticeable that the period necessary for re-establishing an even distribution of the insecticide is shorter under humid than under dry conditions (approximately 4 and 8 days respectively), confirming the greater mobility of the chemical under humid conditions. Blockage of migration due to capillary condensation at high relative humidity did not occur in this case because about 5 days are necessary to saturate the soil layer with water, whereas only 4 days were needed to re-establish the original even distribution of the toxicant in the soil layer.

Experiments under simulated field conditions

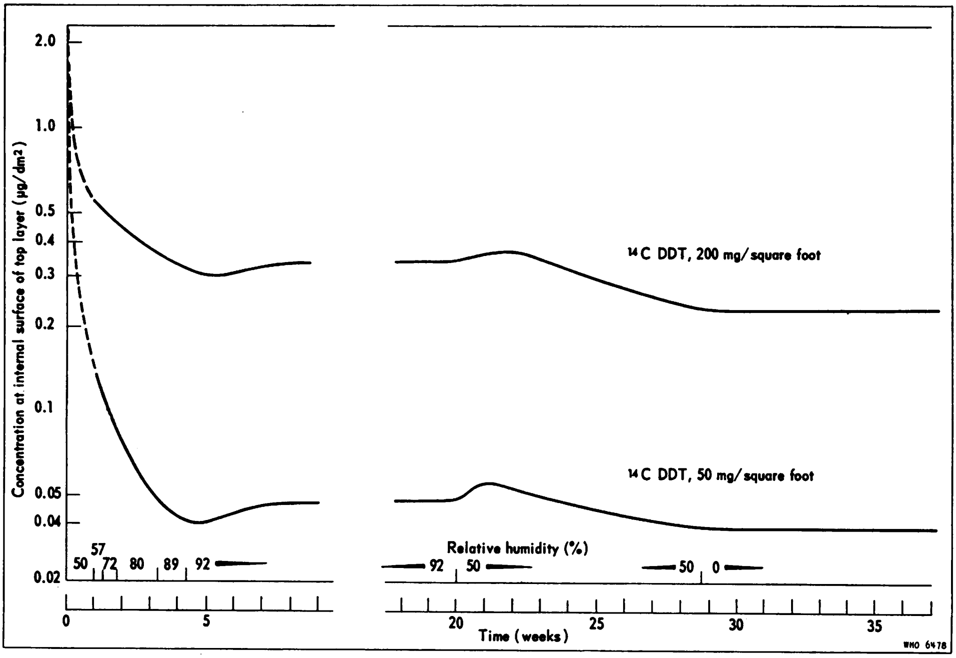
From the evidence gained so far it thus seems conceivable that the rate of inward migration of the sorbed insecticide is not only enhanced with increasing humidity by means of a greater mobility, but that in addition it is directly supported, at least temporarily, by the actual sorption of water (" washing in " effect).

The purpose of the next experiments was to examine the effect of humidity changes on concentration and effectiveness of the insecticide in the top layer of the soil, under conditions simulating as much as possible those in the field.

A thick wall was simulated as follows. Cylindrical blocks of Babati soil were covered all around by adhesive tape except for an area of 3.8 cm^2 for application of the insecticide, the size of which corresponded to that of the window of the Geiger-Müller tube. Metal rings, with an internal cross-sectional area of the same size, embedded in the soil and reaching from the top to about half way

FIG. 4

EFFECT OF CHANGE IN RELATIVE HUMIDITY ON PENETRATION OF ^{14}C DDT INTO SOIL BLOCKS



down the blocks prevented the horizontal spreading of sorbed water and insecticide. The total amount of soil in this experimental set-up represented a column 10 cm high, and as sorption of water vapour will occur on both sides of an actual mud wall, it approximately simulates a wall of 20-cm thickness. The blocks were clamped in holders, enabling easy insertion and withdrawal from the lead-chamber and ensuring that the treated area was always exactly in the same place when measuring radioactivity.

The blocks were first conditioned at 40% relative humidity (temperature 23°-24°C), after which the insecticide was applied. Subsequently, relative humidity was stepped up gradually to approximately 90% in a period of a month, and after that kept constant at that level. It was thought that this would approximately simulate the change from dry to wet season.

As no labelled dieldrin was available at the time of experiments, use was made of ^{14}C DDT. Two blocks were treated, one at a dosage of 200 mg per square foot and one at 50 mg per square foot (2 g/m^2 and 0.5 g/m^2), corresponding to the normal field dosages for DDT and dieldrin respectively. It was assumed that the rate of penetration of these insecticides would not differ appreciably. As only very little ^{14}C DDT was available, it was applied to the surface of the blocks in the form of a homogenized DDT/soil mixture. The first step, i.e., the disappearance of the insecticidal particles from the site of application, is therefore not actually measured, but the dotted lines in Fig. 4 indicate the probable decrease in concentration that would have been found if crystalline insecticide had been applied.

Radioactivity counts were made for periods of 10 minutes for the high dosage and 20 minutes for the low dosage. At the lowest level of activity measured for the 200 mg/square foot dosage the random error was 1.5% (90% probability); for the 50 mg/square foot dosage, 2.6%. Radioactivity was mathematically converted to concentration of insecticide at the internal surface of the soil, the calculations being based on previous tests in which the relation between radioactivity and concentration at the internal surface in homogenized insecticide/soil mixtures had been ascertained.

As can be seen from Fig. 4 there was a rapid inward migration initially, leaving only approximately 0.3 μg and 0.04 μg of the insecticide per dm^2

at the internal surface of the soil in the top layer (0.05 mm) with the high and low dosage respectively. A slight re-migration of the insecticide towards the top layer was noticeable with both treatments. With the 50 mg/square foot dosage it started between 4 and 5 weeks after application, and with the 200 mg/square foot dosage after 5-6 weeks. The probable explanation is that the high rate of " washing in " had led to a temporary building up of a slightly higher concentration of insecticide a little deeper down the soil than in the top layer.

In spite of the high humidity no further inward migration was observed from about 7 weeks onwards, and no change in concentration of insecticide in the top layer occurred so long as the humidity was kept on that high level. It may well be that this is due to capillary condensation of water in the pores of the soil, by which further passage of insecticide molecules is blocked.

A change to 50% relative humidity in the 20th week after application gave, as could be expected, at first a slight increase in concentration (" washing out " effect), followed by a gradual decrease. Apparently, the insecticide regained its mobility, probably because the pores of the soil were no longer blocked, and inward migration proceeded again.

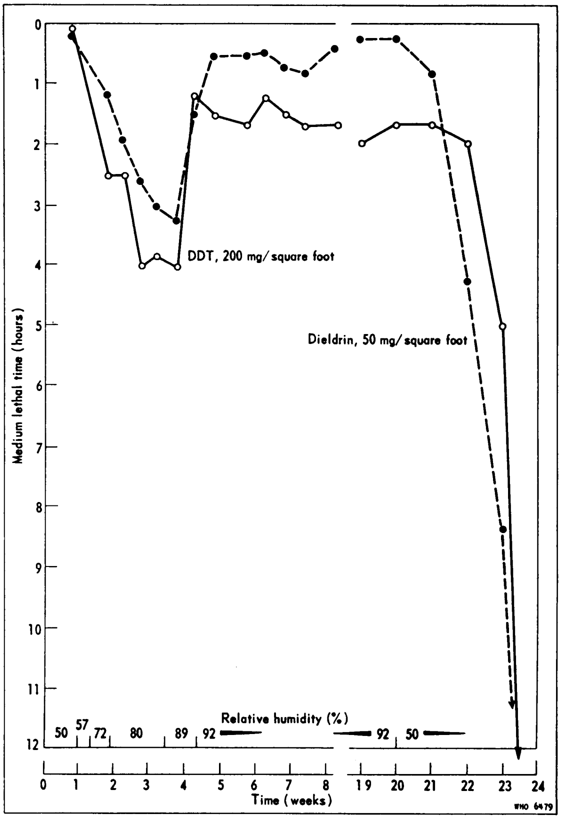
When the relative humidity was lowered still more (to 0 in the 29th week of the experiment) the point was reached where the mobility of the insecticide molecules became hampered again, this time probably because of the strong adsorption forces between insecticide and soil, and the insecticide concentration in the top layer remained constant.

In determining the combined effect of the fall in concentration of insecticide in the top layer and the simultaneous increase in humidity on the biological effectiveness, soil blocks (10 cm high) with a sprayed area of approximately 60 cm² were employed. The surface of the blocks was insulated with adhesive tape except for the sprayed area, thus simulating again a wall of 20-cm thickness. The blocks were sprayed with 50% wettable powders of DDT and dieldrin at a dosage of 200 mg DDT per square foot. and 50 mg dieldrin per square foot respectively. The relative humidity was stepped up as mentioned above. The biological performance of the residues was examined by exposure of 30 female *Aëdes aegypti* mosquitos under glass funnels at regular intervals. The results are given in Fig. 5.

Biological effectiveness decreased at first, and the MLT for *Aëdes aegypti*, i.e., the exposure time required to give a 50% mortality, showed a rapid increase. Thus the influence of the loss in concentration in the top layer of the soil on toxicity was even greater than the influence of the increasing humidity, although the latter factor would normally predominate.

FIG. 5

EFFECT OF CHANGE IN RELATIVE HUMIDITY ON BIOLOGICAL PERFORMANCE OF DIELDRIN AND DDT RESIDUES ON SOIL BLOCKS



The residual effect reached a minimum approximately 4 weeks after application. The relative humidity was then about 90%. Although the blocks contained about half the amount of water they can absorb at that level of humidity, it seemed that the impetus of the " washing in " effect was lost by then, and the toxicity rapidly increased again (decreasing MLT's). Apparently at this stage any further increase in water content of the soil in the top layer did not materially change the concentration of the insecticide any more (capillary condensation), but only improved its availability to the insects.

After about 5 weeks after application both toxicity (which reached a maximum again) and concentration in the top layer (see Fig. 4) did not decrease appreciably. One may expect, therefore, that, providing humidity remained high, this toxic level could be maintained for a considerable period.

An appreciable fall in humidity, however, can be expected to result in a marked decrease in effectiveness. This was shown with the same blocks, when 20 weeks after application the humidity was decreased to 50%. This change in humidity resulted in a very great loss in effectiveness. Five weeks after the transfer to 50% relative humidity no mortality was observed with an exposure period as long as 24 hours.

PROLONGATION OF PERSISTENCE AT LOW HUMIDITY BY ADDITION OF RESINS AND OTHER ADJUVANTS

From the results of the previous experiments it has become clear that dieldrin sorption in soil need not necessarily lead to disappearance of the residual effect provided that the relative humidity remains at an adequately high level. It can be assumed, however, that under conditions of low relative humidity, dieldrin in the sorbed state can lose its activity completely, and then residual toxicity can only be secured by prolonging the persistence of the insecticide in the particulate form on the surface.

It is generally accepted that sorption forces are negligible at distances very little greater than the diameter of a molecule, and thus if one could maintain sufficient distance between the insecticide and the surface sorption might be prevented. It was thought that a promising line of approach might be in the combination of the insecticide with certain types of non-sorbable material, which would thus act as a physical barrier between the insecticide and the sorbent (Gerolt, 1957).

A first sample of such a wettable powder was prepared by mixing equal amounts of powdered dieldrin and powdered coumarone resin, which were then melted together until a homogeneous mixture was obtained. This was cooled and the brittle product finely ground, Teepol being added as wetting agent. The majority of the particles were found to be within the range 10 μ-50 μ. The powder thus obtained was sprayed at a dosage of 1.5 mg dieldrin/dm^2 on to panels of Babati soil. Enough panels were sprayed to allow for a series of tests at successive time intervals, using new panels each time. A standard dieldrin 50% wettable powder, applied at twice the dosage of dieldrin, was included for comparison.

The first results were most encouraging. Table 6 gives a clear picture of the greatly improved persistence as shown in tests with the short-exposure test technique. The standard dieldrin wettable powder gave a 100% kill for a period of one day only on the type of mud used; then mortality figures rapidly fell with time, and the residual effect was completely lost after 2 weeks. On the other hand, the ground dieldrin/coumarone-resin melt showed a high rate of knockdown and 100% mortality throughout the experimental period of 10 months.

No appreciable differences in persistence throughout the test period of 3 months were observed when the ratio dieldrin : resin was altered to 90 : 10, 70 : 30 or 30 : 70. Preliminary experiments with other insecticides (DDT, γ-BHC and aldrin) also showed an improvement as compared with the corresponding wettable powder without resin, but no further work has been carried out along these lines, and experiments have been restricted to dieldrin.

In order to study the possibility of using materials other than coumarone resin for this purpose, a series of adjuvants, incorporated in melts with dieldrin (ratio 50 : 50), were tested for their effect on persistence. Coumarone resin was included in these tests for comparison. The adjuvants were chosen at random except as regards their melting points, which for obvious reasons could not be too far from that of dieldrin. Those examined could be classified into synthetic and natural resins, asphaltous products and crystalline chemicals. The materials tested included coumarone resin, Aroclor 5460 (a chlorinated polyphenyl resin of Monsanto Chemicals Ltd.), Epikote resin (a polycondensation product of Shell, based on epichlorohydrin and bisphenol), colophony, gilsonite (a natural asphaltite), sulfur, dinitro-orthocresol (DNOC), diphenylthiourea, diphenylsulfone and phenylsalicylate. The melts were applied to soil panels at a dosage of 3 mg dieldrin/dm^2 and tested for their persistence, using the short-exposure technique for houseflies.

As shown in Table 7, satisfactory persistence was obtained with all resins (except the Epikote resin), with gilsonite and with sulfur. Their melts with dieldrin remained at an adequate toxic level throughout the testing period of 3 months. The dieldrin/Epikote-resin melt showed a low inherent toxicity, although it was persistent at that level. The dieldrin/DNOC melt lost its originally high toxicity and was was nearly completely de-activated at the end of

TABLE 6

IMPROVED PERSISTENCE OF DIELDRIN/COUMARONE-RESIN SOLIDIFIED MELTS ON SORPTIVE MUD PANELS, EXPRESSED AS PERCENTAGE KNOCKDOWN AND 24-HOUR MORTALITY OF *MUSCA DOMESTICA*

Product	Age of residue	No. of hours after exposure						
		1½	2	3	4	5	6	24
Standard dieldrin wettable powder (3 mg dieldrin/dm²)	2 hours	91	100					100
	1 day	0	3	39	48	52	55	100
	2 days			0	9	17	37	71
	3 days		0	4	4	15	22	56
	1 week					0	2	27
	2 weeks						0	8
Dieldrin/coumarone-resin melt (1.5 mg dieldrin/dm²)	2 hours	49	85	95	100			100
	1 week	0	12	59	94	100		100
	2 weeks	17	72	97	100			100
	4 weeks	14	45	89	100			100
	10 weeks	11	22	44	78	87	98	100[a]
	4 months	4	27	67	87		100	100[a]
	7 months	0	2		52	57	82	100[a]
	10 months		0	13	28	44	56	100[a]

[a] Panels used for exposure more than once.

TABLE 7

BIOLOGICAL PERFORMANCE OF MELTS OF DIELDRIN WITH VARIOUS ADJUVANTS, EXPRESSED AS PERCENTAGE 24-HOUR MORTALITY OF *MUSCA DOMESTICA*

Adjuvant	Age of residue				
	2 hours	2 weeks	1 month	2 months	3 months
Coumarone resin	100	100	100	98	100
Aroclor 5460	100	100	100	100	100
Epikote resin	88	56	63	63	87
Colophony	100	100	100	100	100
Gilsonite	100	100	100	100	100
Sulfur	100	100	100	100	100
Dinitro-orthocresol	100	100	91	71	37
Diphenylthiourea	100	29			
Diphenylsulfone	95	42			
Phenylsalicylate	100	52			

that period. Little or no improvement was observed with diphenylthiourea, diphenylsulfone or phenylsalicylate as additive.

From microscopical examinations of the melts in concentrated suspension in water, it appeared that dieldrin/resin melts were non-crystalline and transparent; only a few needle-shaped crystals were found in the melts with coumarone resin and Aroclor, but the amount of "free" dieldrin was insignificant. Estimation of wettable powder residues on soil panels, by a sampling technique using carboxymethyl cellulose, indicated that the amounts of dieldrin remaining on the surface were high. One month after application the surface residues of dieldrin amounted to 66% of the original deposit with a dieldrin/colophony melt, and with Aroclor the recovery was even higher (Ford—personal communication). In the gilsonite melt a considerable portion of the dieldrin was present in the crystalline form. The melt gave the impression of consisting of gilsonite and dieldrin particles sintered together. A fair amount of "free" dieldrin was found as well, and it was not surprising, therefore, that only onethird of the original amount of dieldrin was prevented from sorption (Hadaway & Barlow, personal communication). The melt with sulfur and DNOC showed a similar structure, although the fragments of dieldrin and adjuvants were in general appreciably smaller than with gilsonite. When wetted by water, the melts with the other crystalline materials tested showed little mutual adherence between dieldrin and the adjuvant.

From the above observations it seems likely that homogeneity is necessary for satisfactory persistence, and that only melts with materials such as resins, having sufficient mutual solubility for dieldrin, secure this property.

Further experiments, conducted both at the Woodstock Agricultural Research Centre and at our associate laboratory in the USA, have shown that the effect of prolonging persistence on sorptive soil surfaces is not peculiar to the resins mentioned above, but that several other synthetic resins have similar activity.

In order to examine to what extent the procedure of melting dieldrin and adjuvant is essential for persistence, a few tests have been carried out with differently prepared combinations, such as simple mixtures obtained by grinding the two components together and evaporation residues obtained from solutions of the components in a common solvent. In all cases a favourable effect on persistence was found, but the product based on melts was distinctly superior. It should be mentioned that with simple mixtures the presence of a filler during grinding prevents any improvement in persistence.

The long persistence of the dieldrin/resin melt was an indication that the transport of dieldrin through the melt and into the soil was very limited. If at the same time, the insecticide from the melt were available to the insects to the same small extent as to the soil, the inherent toxicity of the melt would be much less than that of dieldrin alone.

Tests in which fixed numbers of houseflies were exposed to various dosages of dieldrin alone, 50:50 dieldrin : resin melt, and the corresponding blend showed that the toxicity of the dieldrin is not affected by the addition of coumarone resin. Furthermore, mortalities at a dosage of 0.02 μg per fly were very similar to those obtained with topical application of dieldrin in acetone (LD_{50} for female houseflies$=$0.02 μg/fly), which proves that in all cases the full amount of toxicant was available to the insect.

Epikote resin, on the contrary, greatly reduced the insecticidal activity in the melt (by a factor of approximately 10); this is in agreement with the low insecticidal level of the melt as found on soil panels. Apparently the dieldrin from the melt is not efficiently absorbed by the insects. A similar reduction in toxicity was observed with the dieldrin/Epikote-resin blend, although to a lesser extent.

A full explanation of this phenomenon cannot be given, but it may well be that both dieldrin and resin penetrate the insect cuticle and that the observed differences in inherent toxicity are in some way connected with differences in solubility of the components in, for example, the cuticular wax.

BIOLOGICAL PERFORMANCE OF FINISHED PRODUCTS

In the experiments discussed so far, use was made of hand-ground wettable powders in which no filler had been included. However, no data were available on the performance of finished products based on the principle of the dieldrin/resin melt. Further tests have therefore been carried out with some air-milled wettable powders prepared at the Woodstock Agricultural Research Centre. All products contained 50% dieldrin, 25% adjuvant and 25% filler, plus wetting agents. The adjuvants selected were gilsonite, colophony, coumarone resin and Aroclor 5460, and the dosage applied on to the soil surface was 3 mg dieldrin/dm^2.

Panel exposure tests

A first series of tests was carried out in which 30 adult *Aëdes aegypti* mosquitos (mixed sexes) were exposed to treated soil panels under glass funnels. Exposure tests were done for various lengths of time and at regular intervals up to 3 months after spraying. The panels were stored at low relative humidity (10%) between the tests.

The obvious advantage of this method of testing is that it enables a more precise differentiation in performance between the various products by assessing the minimum exposure time giving a 100% mortality. This principle could not be followed in the housefly tests, as a lethal dose was picked up within 30 seconds with all products. The results of the tests are given in Table 8.

Apart from an initial loss of dieldrin, which is reflected by the difference in toxic effect between the 2-hours and the 1-week tests, all new products showed a remarkable improvement in persistence as compared with the standard dieldrin wettable powder. Apparently, the addition of fillers and the process of air-milling did not affect persistence in this case.

TABLE 8

BIOLOGICAL PERFORMANCE OF WETTABLE POWDERS OF DIELDRIN MELTS WITH VARIOUS ADJUVANTS (PANEL EXPOSURE TESTS), EXPRESSED AS PERCENTAGE 24-HOUR MORTALITY OF *AËDES AEGYPTI*

Adjuvant	Age of residue	Exposure time				
		1 min.	4 min.	16 min.	1 hour	24 hours
None	2 hours	100				
	1 week			7	74	100
	2 weeks			15		100
	3 weeks			0	24	64
	4 weeks			3	6	0
	5 weeks			0	0	14
	6 weeks			0	3	4
	2 months				0	4
	3 months					0
Gilsonite	2 hours	83	100			
	1 week		12	100		
	2 weeks		73	100		
	3 weeks		67	100		
	4 weeks		40	100		
	5 weeks		0	36		
	6 weeks			85	100	
	2 months			97	100	
	3 months			81	100	
Colophony	2 hours	97	100			
	1 week		23	100		
	2 weeks		93	100		
	3 weeks		95	100		
	4 weeks		93	100		
	5 weeks		74	92		
	6 weeks		67	100		
	2 months		55	100		
	3 months		52	100		
Coumarone resin	2 hours	100				
	1 week	83	100			
	2 weeks	67	100			
	3 weeks	74	100			
	4 weeks	82	100			
	5 weeks	13	100			
	6 weeks	41	100			
	2 months	50	100			
	3 months	4	100			
Aroclor 5460	2 hours	100				
	1 week	100				
	2 weeks	100				
	3 weeks	100				
	4 weeks	100				
	5 weeks	91	100			
	6 weeks	90	100			
	2 months	86	100			
	3 months	77	100			

TABLE 9

BIOLOGICAL PERFORMANCE OF WETTABLE POWDERS OF DIELDRIN MELTS WITH VARIOUS ADJUVANTS (TESTS IN EXPOSURE CHAMBER), EXPRESSED AS 24-HOUR MORTALITY OF *AËDES AEGYPTI* AFTER 4 HOURS' EXPOSURE

Adjuvant	Age of residue in months					
	1	2	3	4	5	6
None	3	0	4	0	4	5
Gilsonite	5	0	3	0	10	6
Colophony	3	40	40	27	46	13
Aroclor	92	100	100	93	100	98

Dieldrin/gilsonite was undoubtedly the least persistent mixture of the four. Dieldrin/colophony was slightly more persistent, and dieldrin/coumarone resin was better still. By far the best performance was obtained with dieldrin/Aroclor.

Tests with exposure chambers

The above-mentioned products (except coumarone resin) were also tested in exposure chambers, using the method developed in the US Public Health Service Laboratories at Savannah, Ga. Storage and testing were done at low relative humidity (<50%). Each test was carried out with about 30 female mosquitos, and a fixed exposure period of 4 hours was given. Table 9 gives the results.

It can be seen that under these test conditions, which are undoubtedly more stringent than those of the panel test, only Aroclor gave a satisfactory performance during the entire experimental period. Gilsonite failed, and colophony was intermediate in performance.

TABLE 10

COMPARISON OF RESULTS OBTAINED IN PENAL EXPOSURE TEST AND IN EXPOSURE CHAMBER TEST

Adjuvant	Percentage 24-hour mortality of *Aëdes aegypti*	
	Savannah test (4-hour exposure)	Panel test (4-minute exposure)
None	Nil	Nil
Gilsonite	Nil	Probably nil
Colophony	40	About 50
Aroclor	100	100

By using a box with one plywood panel replaced by glass, it was observed that during a one-hour exposure an individual mosquito walked for an average of approximately one minute only. A comparison between the results of a 4-minute exposure in the panel test and the results of a 4-hour exposure in the Savannah test showed that they tally fairly well. Table 10 gives a summary of results for two and three months after treatment. It can be concluded from this experiment that the mosquitos received the insecticide only during actual walking, and that it was picked up only in the form of particles. Thus, the sorbed part of the dieldrin was not available, which is exactly what could be expected under the low humidity conditions of the tests.

RÉSUMÉ

Les insecticides pulvérisés sur les murs des habitations en boue séchée dans les régions tropicales, perdent plus ou moins rapidement leur activité, en raison d'un phénomène physique de sorption, qui fait disparaître de la surface les particules de DDT ou de dieldrine. Ce phénomène ne s'observe pas seulement avec la boue séchée, mais aussi avec la brique, le plâtre et le papier. Les particules d'insecticide disparaissent rapidement de la surface: plus de 95% d'un dépôt de dieldrine de 0,3 g/m^2 est sorbé par un sol actif en 24 heures. L'expérience montre que l'humidité est un facteur primordial du phénomène de sorption, et l'auteur l'a particulièrement étudié.

Les essais ont été faits avec des sols expérimentaux et du DDT, en partie marqué par du ^{14}C, ainsi que de la dieldrine, selon des techniques décrites en détail par l'auteur.

De ses résultats, l'auteur conclut que la sorption ne nuit à l'efficacité des insecticides que dans des conditions d'humidité faible ou moyenne. Dans des conditions de forte humidité, la concentration superficielle de l'insecticide et ses chances d'action sur les insectes ne changent pas. On admet que la condensation capillaire de l'eau dans les interstices du sol empêche la migration des particules vers l'intérieur. Les forces de sorption ne s'exercent guère qu'à des distances équivalant au diamètre d'une molécule, et le problème, pour remédier à l'effet de la sorption, consiste à maintenir une distance de cet ordre de grandeur entre l'insecticide et la surface sorbante. On a cherché à le faire en combinant l'insecticide à un matériau non sorbable, qui jouerait le rôle de barrière physique. Une préparation a été mise au point à cet effet, combinant la dieldrine en poudre à des adjuvants (résines ou asphaltes), le tout étant fondu jusqu'à homogénéité. Après refroidissement, la masse est réduite en poudre, dont les particules ont 10-50 μ. Ce mélange est pulvérisé sur des surfaces, à raison de 1,5 mg/dm^2. Les résultats ont été très encourageants. Alors que sur une surface, faiblement humide ou sèche, la dieldrine perdait tout pouvoir en deux semaines, la dieldrine/résine n'avait rien perdu du sien après 10 mois. La résine agit de la même façon avec le DDT, le HCH-gamma et l'aldrine.

Les résultats les meilleurs sont obtenus avec les résines et les insecticides mutuellement solubles. Mais certaines, telle la résine Epikote, abaissent la toxicité intrinsèque de l'insecticide. Des produits divers associés à la dieldrine — Gilsonite, colophane, résine coumarone et Aroclor 5460, selon diverses techniques d'exposition, ont tous agi dans le même sens, dans l'ordre précité d'efficacité croissante, l'Aroclor étant de loin le meilleur.

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