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RESIDUAL INSECTICIDES AND THE PROBLEM OF SORPTION

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SYNOPSIS

Whereas laboratory investigations have elucidated the mechanism of sorption of residual insecticides and demonstrated that their persistency is determined by a number of physico-chemical factors and is therefore theoretically calculable, the variables encountered in the field may produce results in apparent conflict with those theoretically expected. Attempts to enhance persistency through the prevention of sorption, although promising, have so far not been fully successful. It is consequently also necessary to assess the residual effectiveness of insecticides, " effectiveness " here being viewed as a biological effect expressed in terms of the mosquito mortality produced. For this purpose bio-assay tests have been used, but with very variable results, and it is suggested that a study of the bio-assay technique itself is needed. This should be conducted in parallel with chemical determinations of the total amount of insecticide present both on and below the sprayed surface.

The value of residual insecticides employed for malaria eradication or control depends on the vector mortality maintained after their application. This is a biological effect which can be evaluated by various entomological methods including bio-assays. From the viewpoint of malaria eradication or control the effectiveness of the residual insecticide is expressed by the reduction of the mean life expectation of the vectors to such a level that transmission of the infection is rendered impossible. This will depend on the following main factors:

1. Intrinsic tolerance level of the insect (specific toxicity)
2. Toxicity of the insecticide and its mode of action
3. Bionomics of the mosquito
4. Maintenance of the toxic dose = *persistency* (insecticide alone)

effectiveness (insect-insecticide relationship)

This paper deals essentially with those factors which, after the insecticide is applied, affect the *persistency* of insecticide deposits and thus influence its *effectiveness*.

Persistency and Effectiveness : Definitions

Most authors speak of the residual effect of an insecticide in terms of " persistency " or " effectiveness " without differentiating between the two. These two terms, however, are not really identical and to distinguish between them is necessary not only for reasons of etymology, but even more pertinently for practical reasons.

Etymologically, " persistency " means " steadfast continuance " (*persistere*); the term " persistency " should be employed to mean the amount of an insecticide which is present at any given time, either on or below the treated surface, whether in an active or in a potentially active form. With this connotation, *persistency* expresses a physico-chemical fact which can be directly determined only by physico-chemical means ; it is conditioned only by factors extrinsic to the insect.

On the other hand, the term " effectiveness " should be restricted to the biological effect resulting from the application of insecticide deposits, from the amount of insecticide present; that is, it should be restricted to the expression of the mosquito mortality produced, the over-all biological effect being conditioned by intrinsic factors of the mosquito and its ecology.

Physico-chemical Factors and Sorption

The persistency of an insecticide depends mostly on two factors: the nature and formulation of the insecticide on the one hand, and the nature and type of the surface on the other.

The nature and formulaton of the insecticide

Essential physico-chemical characteristics which determine the persistency of a given insecticide are vapour pressure and volatility. Table 1 gives vapour pressures for DDT, dieldrin and gamma-BHC (Brown, 1951; de Ong, 1956; Gunther & Blinn, 1955).

Perusal of Table 1 shows that the vapour pressure of DDT is slightly greater than that of dieldrin at room temperature, and that each is about sixty times less than that of gamma-BHC.

Table 2 shows that the rate of volatilization is higher for dieldrin than for DDT below about 43°C, while it is the reverse above this temperature.

These figures, though obviously not directly applicable for forecasting the persistency of formulations under field conditions, should be kept in mind as giving the trend and the order of persistency of these insecticides, since theory agrees with observations to show that the higher their values the shorter will be the persistency.

Barlow & Hadaway (1958a) have measured the persistency of DDT, dieldrin and gamma-BHC on glass plates and other impervious or moderately pervious materials. Their figures for the evaporation of gamma-BHC and dieldrin from glass plates at 25°C and approximately 60% relative humidity are as follows :

Gamma-BHC		*Dieldrin*	
Time (weeks)	*Average weight recovered (g/m^2)*	*Time (weeks)*	*Average deposit (g/m^2)*
0	1.06	0	1.00
1	0.50	11	0.84
2	0.06	24	0.50
4	0.0	52	0.10

TABLE 1. VAPOUR PRESSURES OF THE THREE MAIN CHLORINATED-HYDROCARBON INSECTICIDES

Insecticide	Vapour pressure (mm Hg)		
	20°C	25°C	40°C
DDT	1 to 1.5×10^{-7}	3×10^{-7}	3×10^{-6}
Dieldrin		1.8×10^{-7}	
HEOD	1.0×10^{-7}		1.3×10^{-6}
Lindane (gamma-BHC)	9.4×10^{-6}	3.0×10^{-5}	

TABLE 2. RATE OF VOLATILIZATION OF ALDRIN, DIELDRIN, LINDANE AND DDT *

Aldrin		Dieldrin		Lindane		DDT	
temperature (°C)	rate (g/cm^2/sec.)	temperature (°C)	rate (g/cm^2/sec.)	temperature (°C)	rate (g/cm^2/sec.)	temperature (°C)	rate (g/cm^2/sec.)
32.2	3.39×10^{-7}	20.4	8.17×10^{-9}	20.4	1.16×10^{-7}	28.4	2.75×10^{-9}
27.2	4.95×10^{-7}	28.4	1.66×10^{-8}	28.4	2.84×10^{-7}	33.5	6.52×10^{-9}
28.2	3.54×10^{-7}	33.5	1.99×10^{-8}	33.5	5.78×10^{-7}	38.6	1.47×10^{-8}
39.6	8.85×10^{-7}	40.8	3.52×10^{-8}	40.8	2.27×10^{-6}	40.8	2.01×10^{-8}
45.7	7.76×10^{-7}	54.0	7.51×10^{-8}			44.4	4.71×10^{-8}
		59.3	1.08×10^{-7}			48.8	1.04×10^{-7}
		65.2	2.57×10^{-7}			54.0	2.26×10^{-7}
		69.8	2.34×10^{-7}			59.3	4.33×10^{-7}
		75.8	3.62×10^{-7}			65.2	9.72×10^{-7}
		83.2	6.15×10^{-7}			69.8	1.67×10^{-6}
						75.8	3.64×10^{-6}

*** Reproduced, by permission, from *Rate of volatilization of aldrin, dieldrin, lindane and DDT* (Shell Agricultural Bulletin ADB: 199)**

TABLE 3. DOSAGES OF DDT AND DIELDRIN IN MG PER SQUARE FOOT * ON VARIOUS SURFACES SPRAYED WITH 0-10 MICRON CRYSTALS AT 50 MG PER SQUARE FOOT KEPT AT 25°C AND 60%-70% RELATIVE HUMIDITY **

Surface	Time after spraying (months)					
	0		3		6	
	DDT	dieldrin	DDT	dieldrin	DDT	dieldrin
Glass	49	54	—	—	49	19
Bamboo	47	49	—	—	47	25
Palm leaf	47	45	—	—	42	16
Wood, surface deposit	31	27	34	17	32	13
Wood, first layer	16	18	18	10	19	9
Wood, second layer	0	0	0	0	1	0

* A convenient conversion rate is 100 mg per square foot = 1 g per square metre.

** Reproduced, by permission, from Hadaway, A. B. & Barlow, F. (1957) Unpublished document CIRU/Porton/Report No. 139 (Colonial Insecticides Research Unit, Porton Down, Wilts, England)

These results, and those given in Table 3, are in general agreement with those in Tables 1 and 2.

As regards the results in Table 3, it was evident upon microscopical examination that after three months no striking losses had occurred from the glass, bamboo and palm leaf other than loss of dieldrin by evaporation, and that is why no analyses of the residue were made. On the wood, DDT had not changed in either amount or distribution, while the dieldrin had suffered some loss, apparently by evaporation, but had not diffused under the surface.

After six months there was little change in the DDT whereas a considerable amount of dieldrin had evaporated from all the surfaces. The average loss, about 66%, agrees fairly well with a previous determination on glass where the loss in six months was 50%.[1]

Thus at the dosages used no DDT was lost, whereas dieldrin steadily evaporated so that about 40% remained after six months. Under laboratory conditions, commercial formulations of DDT, dieldrin and BHC follow the same order and range of persistencies as pure preparations (Woodruff & Tuner, 1947).

Under field conditions, the number of possible variables influencing an experiment and the inherent technical difficulties may sometimes substantially alter the theoretical picture unless trials are made on significant numbers. Careful analysis will, however, make it possible to determine the variables,

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[1] Barlow, F. & Hadaway, A. B. (1957) Unpublished document CIRU/Porton/Report No. 135 (Colonial Insecticides Research Unit, Porton Down, Wilts, England)

assess their respective influences and show the consistency of results with those obtained in the laboratory. This is the explanation for some apparently conflicting results obtained by Langbridge [1] at Birnin-Kebbi, Northern Nigeria. In comparative trials with 50% water-dispersible powders applied on thatch, the chemical estimation gave figures for the average loss, in mg per day per m², of 2.4 for dieldrin, 5.0 for DDT, and 20.0 for gamma-BHC, giving a ratio of persistency of roughly 1 : 2 : 10, while from the ratio of their vapour pressures one would expect for DDT a persistency of approximately sixty times that of gamma-BHC. But the author correctly points out in a personal communication (1959) that the loss of DDT and dieldrin was calculated over 65 days whereas the loss of BHC was calculated over only 3 days as no BHC was detectable on the surface after that period. If persistency is calculated in relation to the time of disappearance of half the deposit, the ratio BHC : dieldrin : DDT is 1 : 25 : 70, which agrees with the order of vapour pressure. The difference between DDT and dieldrin is attributable to the greater amount of DDT present. It is also interesting to note that with gamma-BHC the percentage of insecticide recovered (from impervious surfaces) does not seem to be much influenced by the dosage applied: Langbridge noted that the amount present between 5 and 16 weeks after spraying is approximately 0.1 g/m², whether the initial dosage was of 0.4 or 0.8 g/m², while Duerden, quoted by Langbridge in the document cited, has shown that at dosages of 0.1, 0.2 and 0.4 g/m² only 6% of the three deposits were recoverable after six days.

On the whole, therefore, field results confirm laboratory data and show that persistency of DDT, dieldrin and BHC on impervious surfaces is directly related to their vapour pressures and volatilities. Among the factors which may induce variations or apparent deviations from the expected order of persistency two in particular should be considered. The first are climatic factors, such as temperature, relative humidity and ventilation. The second are physical factors, which may be divided into, first, the type of surface (irregular or corrugated) where the effective surface area may be much more than the measurable plane surface, with consequent reduction in the dosage per unit area of surface and increase of evaporation; and secondly, the formulation in the case of the so-called " resin " mixtures in which the resin acts as an extender by slowing down the release of insecticide through volatilization while the specific vapour pressure remains unchanged.

Wettable powders

In addition to vapour pressure and volatility, persistency of insecticides is influenced by the physical form in which they are used: dust, solution, emulsion, paste, or water-dispersible powder. In most malaria eradication campaigns insecticides are used as water-dispersible (wettable) powders;

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[1] Unpublished document WHO/Mal/Inform/32, March 1958

chlorinated insecticides in this form are compounded with a diluent clay, but in the later stage of preparation various wetting, suspending and anticaking agents are added, as shown in Table 4.

TABLE 4. COMPOSITION AND PHYSICAL CHARACTERISTICS OF VARIOUS INSECTICIDE WATER-DISPERSIBLE POWDER CONCENTRATES, CONFORMING TO INDIAN STANDARD SPECIFICATIONS *

Property tested	Water-dispersible powder concentrate			
	50% DDT	75% DDT	50% BHC	50% dieldrin
Composition				
Insecticide content, %	48.9	74.9	49.8	49.9
Clay (inert carrier) content, %	41.7	14.9	40.2	40.0
Dispersing agents content (water-soluble constituents), %	9.4	10.2	10.0	10.1
Physical properties				
Suspensibility, %	64.8	73.7	56.3	64.6
Suspensibility after accelerated tropical storage, %	56.6	60.1	54.8	58.4
pH	9.2	7.0	7.0	7.0
Particle size of the active toxicant: Below 74 microns, % **	99.4	100.0	100.0	100.0
Below 40 microns, % †	96.0	98.0	97.0	95.0
Below 20 microns, % †	83.0	90.0	80.0	85.0
Below 10 microns, % †	65.0	72.0	63.0	63.0

* Reproduced, by permission, from Bami (1958)

** Results of sieve analysis

† Calculated from data presented in Bami's paper

Wettable powders are the most suitable formulations for use in malaria campaigns not only for practical and economic reasons but also for technical reasons, among which is their longer persistency on pervious surfaces. Although even water-dispersible powders may undergo sorption, they are liable to persist longer on sorptive surfaces than either solutions or emulsions, which upon application penetrate rapidly under the surface. The aqueous suspensions leave the insecticide and the fillers on the surface even though the water is absorbed within the interior of the wall. Further penetration of the insecticide takes place only gradually and depends on the sorptive capacity of the surface. Inert diluents of water-dispersible powder have only a slight effect in reducing the speed of sorption of the insecticide, as has been shown by Hadaway & Barlow (1952b).

Particle size of aqueous suspensions

In view of the observations made previously, it is clear that as persistency is related to vapour pressure and volatility, the larger are the crystals of the insecticide the longer will they persist. A number of observations of Hadaway & Barlow (1952b) show that this is so on impervious as well as on pervious surfaces. On impervious surfaces persistency is essentially controlled by the above-mentioned factors and therefore it is directly influenced by temperature, relative humidity and ventilation. On pervious surfaces persistency is also affected by the specific sorptive power of the wall surface and will depend on the interplay of all these factors. On mud blocks at 25.5°C and at a relative humidity of 50%, particles in the 0-10 micron range were no longer visible two days after application; those in the 10-20 micron range had disappeared by the end of the first week, and those in the 20-40 micron range by the end of the second week. These authors (1952c) have also shown with *Aëdes aegypti* that for DDT there is an inverse relationship between particle size and effectiveness and that the optimum particle size of crystals for anophelines is from 0 to 10 microns.

Hadaway & Barlow's results agree with the observations of the majority of authors (Smith & Goodhue, 1942; Woodruff & Tuner, 1947; Kido & Allen, 1947) and especially with those of Roth, Kocher & Treboux[1] as well as of Bami et al. (1958), who have shown recently that in standard formulations at least 50% to 70% of the insecticide contents are likely to have a particle size below 10-20 microns (see Table 4).

While the small particle size is a definite advantage with regard to biological effectiveness (on account of the ease with which mosquitos pick up the toxicant), it is an inconvenience with regard to persistency on pervious surfaces, since not only are small particles sorbed much more rapidly, but on account of their greater surface per weight unit they are also much more rapidly volatilized. This conflicting situation increases the difficulty in formulating water-dispersible powders which must be both biologically efficient (and therefore contain a high percentage of small crystals in the range 0-10 microns) and highly persistent. Effectiveness would require a high percentage of small crystals in the range 0-20 microns; a high persistency would require rather coarse particles. The right solution is probably a compromise.

The type of surface

Surfaces met with during spraying campaigns may be roughly distinguished into sorptive and non-sorptive.

Non-sorptive or *impervious surfaces* are, for instance, concrete (depending on its composition), wood, bamboo, banana leaf, grass, etc.; thatched

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[1] In a paper entitled *Particle size and effect for DDT wettable powders* presented at the Fourth International Congress for Crop Protection, Hamburg, September 1957.

roofs are typical examples. The persistency of the insecticide on these surfaces is conditioned mainly by the physico-chemical characteristics of the formulations.

Sorptive or pervious surfaces are most frequently represented by mud walls (adobe). There is quite a variety of such surfaces depending on the type of mud (laterite, clay, loam, etc.) and the extraneous material—either originally present or intentionally mixed with the mud to improve its building qualities (grass, wood, sand, stones, cow dung, etc.). On such surfaces the persistency of insecticide deposits will depend not only on the nature of the insecticide or its formulation, but also on the nature and structure of the mud.

Observations in the laboratory and in the field show that on all muds the insecticides disappear in the order BHC, dieldrin, DDT, and at a speed related to their respective rates of diffusion in the mud and approximately related to volatility (Hadaway & Barlow, 1952a, 1952c; Burnett, 1957).

A brief outline of the fundamental process of sorption may be useful at this point.

The Fundamental Process of Sorption

In 1955, Barlow & Hadaway suggested that sorption of insecticides took place from the vapour phase. In recent research the same authors have shown that " the vapour phase is only of minor importance in the transfer of insecticide from particles to the active surface of the mud ", and that " by far the greatest movement must be brought about by diffusion directly from the crystals over the surface of the soils; this is then followed by further diffusion inside the mud layers " (Barlow & Hadaway, 1958a). The rates of diffusion of BHC, dieldrin and DDT are roughly of the same order as their volatilities and are influenced by temperature and relative humidity.

Temperature

An increase in temperature gives an increased rate of sorption (Barlow & Hadaway, 1955). This influence is greater on the first stage—that is, on the speed of movement of the insecticide molecules from crystals lying on the surface to the superficial layer—than on the second stage of diffusion inwards (Barlow & Hadaway, 1958a).

Relative humidity

Following field observations by Hocking (1947), E. Bordas & L. Navarro [1] and Burnett (1957), increases in the relative humidity were seen to be followed by an increase in the mortality of mosquitos. Hadaway and

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

Barlow have given an experimental demonstration and an explanation of the phenomenon (Hadaway & Barlow, 1956; Barlow & Hadaway, 1958a). In particular they were able to make comparative observations on persistency and effectiveness.

Influence on persistency

As relative humidity increases, the rate of initial sorption decreases for gamma-BHC, dieldrin and DDT. For instance, on highly sorptive Uganda mud blocks at 90% relative humidity (RH), gamma-BHC and dieldrin, though more persistent than at 10% RH, disappeared in a few days. At 10% RH, DDT disappeared in one to two days, at 50% RH in five to six days, and at 90% RH it remained as a surface deposit for more than seven weeks. Thus, the influence appears to be much greater on DDT than on the other two toxicants. On less active soils, dieldrin also has a very long life at 90% RH compared with that at low relative humidities.

Once the insecticide has been sorbed within a narrow superficial layer of mud, further diffusion inwards is directly related to humidity, and a decrease of concentration in the surface layers tends to take place.

As to the phenomenon of desorption, there is no apparent movement of sorbed insecticide back towards the surface irrespective of the relative humidity until the concentration of insecticide is approaching uniformity throughout the mud wall. Only DDT has been noted to show a tendency to desorption before a uniform concentration is reached and only at 90% RH. Uniform concentration depends on the rate of diffusion and thus is reached much more rapidly with gamma-BHC than with dieldrin and DDT. Desorption after uniform concentration is reached has been marked only for gamma-BHC, while for dieldrin and DDT it was negligible even after 12 months.

Influence on effectiveness

An increase in humidity is followed by an increased effectiveness of insecticide deposits, as far as its contact and fumigant action are concerned.

This change in activity of the deposits is reversible and a reduction in humidity is followed by a reduction in mortalities of mosquitos entering the huts or in contact with the sprayed wall.

The general increase in biological activity of sorbed insecticides at high humidities is due not to an increase in concentration in the surface layers of the mud but to a probably greater activity of insecticide molecules and to their greater availability to mosquitos, even at lower concentrations.

Tables 5, 6 and 7 show the effect of changes in relative humidity on mortalities.

The practical implications of these observations for field work have been described as follows by Barlow & Hadaway (1958a):

TABLE 5. EFFECT OF CHANGES IN RELATIVE HUMIDITY UPON MORTALITIES OF BLOOD-FED FEMALES OF *AËDES AEGYPTI* EXPOSED DIRECTLY TO MUD BLOCKS TREATED WITH GAMMA-BHC *

Relative humidity conditions	Percentage kill 24 hours after exposure for following number of minutes:				
	2	4	8	16	32
90% for 24 hours	14	65	95	100	
35% for 24 hours			0	0	0
90% transferred to 35% for 24 hours				0	0
35% transferred to 90% for 24 hours	0	35	85	100	

* Reproduced, by permission, from Barlow & Hadaway (1958b)

TABLE 6. EFFECT OF RELATIVE HUMIDITY UPON MORTALITIES OF BLOOD-FED FEMALES OF *AËDES AEGYPTI* EXPOSED TO DDT-TREATED MUD BLOCKS *

Relative humidity conditions	Percentage kill 24 hours after exposure for following number of minutes:	
	64	128
90% for 48 hours	13	100
40% for 48 hours	0	0
90% transferred to 40% for 48 hours	0	0
40% transferred to 90% for 48 hours	8	98

* Reproduced, by permission, from Barlow & Hadaway (1958b)

TABLE 7. TOXICITY TO BLOOD-FED FEMALES OF *AËDES AEGYPTI* OF DIELDRIN-TREATED UGANDA MUD BLOCKS KEPT AT DIFFERENT HUMIDITIES *

Humidity conditions	Time	Percentage kill 24 hours after exposure for following number of minutes:						
		2	4	8	16	32	64	128
90%	12 weeks		5	55	98			
Transferred to 30% for 24 hours							0	0
30%	12 weeks					0	20	63
Transferred to 90% for 24 hours		5	28	75	100			

* Reproduced, by permission, from Barlow & Hadaway (1958b) .

" In the field at any given moment, the toxicity of a mud surface sprayed with an insecticide will depend upon humidity in two ways. In the first place the humidity conditions during the time between spraying and testing will determine how much of the insecticide is sorbed and, of the fraction that is sorbed, how much will remain in the layers of mud nearest to the sprayed surface. Assuming that sorption is complete, the concentration of insecticide in the mud will then give a kill of insects which is controlled by the humidity at the time of exposure. For example, if dieldrin is sprayed on to an active soil, all will be sorbed in a few days, rapidly at low humidity, more slowly at high humidity. If the low humidity persists, diffusion of dieldrin deeper into the mud will be slow, whereas at high humidity it will be fast. At any given time the biological activity, determined at the humidity at which the mud has been kept, will be less at low humidity than at high because the greater potency at high humidity during the test period will more than offset the decreased concentration of insecticide in surface layers. However, if the humidity suddenly increases, the biological effectiveness of the mud will be greater than if it had been kept under high humidity conditions from the beginning. This, of course, is because of the greater concentration of dieldrin in the mud near the insects. In the same way, if a mud which has been in a high humidity is moved to a low humidity it will be less effective than if it had been kept at the same low humidity all the time because the concentration of dieldrin in the surface layers of this mud would be lower."

Sorption of Gamma-BHC

The persistency of gamma-BHC on the application surface is much less than that of dieldrin or DDT on either sorptive or non-sorptive surfaces. However, for gamma-BHC sorption is an advantage, as in a sorbed condition it is slowly released from the mud; but desorption and evaporation take place only after the concentration of the insecticide is fairly uniform throughout the depth of the wall. As this is dependent upon diffusion, which in turn depends on humidity, the residual effectiveness of gamma-BHC on mud will depend also on relative humidity. At a high humidity a high biological activity for a relatively short time would be expected, whereas at a low humidity a low biological activity for a much longer time would be expected.

In field experimental huts in Tanganyika, Burnett (1957) has shown that on a highly sorptive mud a dose of 0.20 g/m^2 of gamma-BHC from a 50% water-dispersible powder produced mortalities of approximately 70% with *A. gambiae* and *A. funestus* for up to nine months. On a poorly sorptive surface (banana-leaf) and a moderately sorptive surface (mud and sand) mortality fell down to 20% after 4 months and 42% after 7 months respectively.

Practical influence of relative humidity on insecticides under field conditions

The extent to which relative humidity affects the persistency and the practical effectiveness of insecticides under field conditions has not been yet the subject of enough investigations to allow of any definite conclusions. Observations carried out so far under controlled conditions by Bordas &

TABLE 8. RANGES OF VARIATIONS OF RELATIVE HUMIDITY AND TEMPERATURE INSIDE AND OUTSIDE AN AFRICAN HUT AT MAGUGU, TANGANYIKA *

Month	Relative humidity (%)				Temperature (°C)			
	outside		inside		outside		inside	
	maximum ranges	minimum ranges	maximum ranges	minimum ranges	maximum ranges	minimum ranges	maximum ranges	minimum ranges
March	86-93	14-50	83-98	44-78	25.6-33.3	15.6-20	25.6-28.9	20-22.2
April	86-94	18-54	88-99	52-84	26.7-31.1	15.6-20	23.3-30	18.9-22.2
May	88-94	37-60	83-98	62-86	24.4-30	13.3-18.9	24.4-27.8	15.6-21.1
June	83-92	18-69	77-97	54-90	22.2-28.9	10.5-17.8	21.1-27.8	14.4-20.0
July	78-92	2-56	74-90	54-77	21.1-28.9	9-16.7	20-26.7	13.3-17.8

*** Based on figures from van Tiel, N., *Field and laboratory experiments on the performance of dieldrin wettable powder on sorptive surfaces* (unpublished interim report issued by the Shell Co., 1958)**

Navarro,[1] by Burnett (1957) and by van Tiel [2] clearly show that the increase in relative humidity tends to compensate for the consequence of sorption of chlorinated insecticides, particularly dieldrin. But if this is enough to confirm the validity of the principle, it does not suffice to determine its range and extent. Here again, more investigations are needed.

There is evidence to show that in most types of African hut relative humidity is fairly high and stable. Hadaway & Barlow (1952b) quote the climatic conditions recorded by Haddow (1942) for an African hut with mud walls and grass roof at Kisumu, Kenya. This hut was a well-insulated structure, the temperature indoors being about half that outdoors with a mean daily minimum of 20°C and a mean maximum of 33°C; the relative humidity ranged from 38 % to 79 % on dry days and from 57 % to 83 % on wet days. More recently, van Tiel [2] at Magugu, Tanganyika, has reached the same conclusions. It appears from Table 8, which is based on his figures, that even during the dry season (from 15 May onwards) the minimum relative humidity figures inside the hut investigated by him rarely fell below 50 %, the average relative humidity in the hut being well over 60 %.

It is obvious, however, that the particular conditions of relative humidity and temperature of a few huts cannot be expected to reflect the general picture of climatic conditions prevailing in an area. Temperature and relative humidity inside houses or huts are themselves dependent on a number of factors: physiography and meteorological conditions, local housing traditions (material used for construction, size and number of

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

[2] *Field and laboratory experiments on the performance of dieldrin wettable powder on sorptive surfaces* (unpublished interim report issued by the Shell Co., 1958)

openings, etc.), and local customs (cooking, ventilation, etc.). Moreover, the relative humidity and temperature of the wall itself are different from those of the environment. Thus the extent of the favourable effect exercised by relative humidity on insecticides is controlled by many local factors rather difficult to assess accurately; but all observations point to the conclusion that during the wet season in tropical and subtropical areas the effectiveness of insecticides is improved. It can safely be stated that in the field high relative humidity will enhance effectiveness.

Proportion of sorptive and non-sorptive surfaces

In view of the striking differences in persistency observed between insecticide deposits on pervious and impervious surfaces, the proportion of each type of surface should be known in any area where residual spraying must be performed.

Langbridge[1] in Northern Nigeria noted that on the average 60%-70% of the sprayable surface in local villages was of thatch while 30%-40% was of mud, and he showed that the persistency on these two different materials varied a great deal. For BHC, for instance, six days after spraying on thatch an average of only 16% of the initial deposit could be recovered, while on mud 30% could be recovered if the initial deposit was of 0.4 g/m^2 and 50% if the initial deposit was of 0.8 g/m^2.

The problem of the persistency of DDT and dieldrin is analogous, the difference being only in the rate of disappearance.

Nature of the mud

Granted that sorption takes place by a physical process of diffusion (Barlow & Hadaway, 1958a), the degree and speed of sorption varies with the nature and the constituents of the mud. A number of studies have been devoted to the fundamental process to determine the responsible factors.

Chemical factors

Downs, Bordas, & Navarro in 1951 expressed the opinion that, with DDT, inactivation might be due to the chemical degradation of DDT to DDE through dehydrochlorination, owing to the presence in the soil of oxides of iron and aluminium. In spite of a number of further studies and although Bordas & Navarro still maintained in 1955[2] that inactivation occurs through a chemical process, their opinion was not corroborated by any further evidence. This is clearly borne out by the studies of Barlow & Hadaway (1955) and Alessandrini et al. (1956).

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[1] Unpublished document WHO/Mal/Inform/32, March 1958

[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)

Langbridge[1] has shown that the sorptive capacity increases with decreasing particle size of muds and that the percentage of iron and aluminium oxides is higher as the particle size decreases. The apparent correlation between the inactivating properties of muds and their iron oxide content could be explained if in the process of weathering the iron oxide and alumina constituents were ground down preferentially into fine particles having a large specific surface.

The higher inactivating properties of muds with metallic oxides would be therefore rather a coincidence than a cause.

In a recent study, Press (1959) has systematically measured adsorption of insecticides on mud-block samples from Zanzibar, Pemba and Taveta Pare, Tanganyika, using the flowing chromatography method after the insecticide had been dissolved in petrol ether. While these experimental conditions are not those met with in routine insecticide spraying, the conclusions reached are interesting:

(1) a high aluminium and iron content together with a low silica content is coincident with an increase in the adsorption properties; this in the author's opinion suggests that there is a relationship between the chemical constitution of muds and adsorption;

(2) there is a relationship between the chemical constitution of the insecticides and adsorption, following roughly this decreasing order of adsorptive properties:

acids and bases
alcohols, thiols
aldehydes, ketones, epoxides (e.g., dieldrin)
halogen-containing substances (e.g., BHC and DDT), esters
unsaturated hydrocarbons
saturated hydrocarbons.

Accordingly the author has noted that dieldrin is more adsorbed than gamma-BHC and DDT, both in alumina (the adsorbent used for comparison) and in soil samples of differing adsorption strength.

These observations are enlightening since they bring out the interplay of the chemical constitution of muds and of that of insecticides. The experimental conditions were here carefully standardized, thus underlining by contrast the complexity of the phenomenon and of its variations under natural conditions.

In another recent work, Barlow & Hadaway (1958a) have shown with Uganda soil that removal of all materials soluble in organic solvents and removal of iron compounds had no influence on the rate of sorption of dieldrin and only a slight limiting effect on the sorption of DDT.

The same authors were also able to prove that sorption of dieldrin proceeds without decomposition even on soils of high surface acidity; on

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[1] Unpublished document WHO/Mal/Inform/32, March 1958

the other hand, neither by qualitative colour tests, nor by paper chromatography nor by crystallization of the recovered insecticides could they find evidence of any chemical decomposition.

From the above studies it would appear that: (*a*) chemical degradation of insecticides does not take place on mud surfaces to any significant extent; (*b*) the chemical constituents of the mud may sometimes exert an influence on sorption; and (*c*) the chemical composition of insecticides has a direct bearing on sorption.

Physical factors

In 1952, Hadaway & Barlow suggested that the sorption of BHC took place from the vapour phase and assumed the same mechanism in the cases of DDT and dieldrin. In 1955, they were able to confirm that the loss of insecticide occurs by adsorption.

Further studies by the same authors and others confirmed that inactivation is essentially attributable to a physical process of adsorption, not so much from the vapour phase as by diffusion (Barlow & Hadaway, 1958a). The majority of studies have since tried to elucidate the physical factors involved in this process, among them the following.

Particle size of muds. In 1956, Paulini, using the carbon-tetrachloride technique proposed by Barlow & Hadaway (1955), showed that the sorptive capacity of muds depends directly on the proportion of the colloidal fraction in the mud (particles 0-60 millimicrons in diameter).

Surface area of muds. Langbridge [1] came to the conclusion that the inactivation of insecticides by mud walls is due to the large surface area per unit weight of the mud and that with increasing surface area the sorptive capacity increases and the persistency of DDT decreases.

Confirmation of the direct relationship between the particle size and sorption is also shown by the sorption of methylene blue from an aqueous solution on the sub-60-millimicron fraction of three muds of Northern Nigeria. Langbridge's results appear in Tables 9 and 10 and are comparable with those obtained using DDT.

Small differences may be noted between the surface areas given in Tables 9 and 10, but the general trend of sorption as related to the surface area is maintained.

In sorbing methylene blue the three building muds behave identically with sorbents such as animal charcoal, which is classically known to exhibit physical (van der Waals) adsorption forces. Press's results (1959) also confirm that finely particulate soils are relatively more sorptive.

Thus, as theory suggests, these observations tend to prove that sorption is related to the specific surface area of the particles in the mud, particularly those of larger specific area by unit, i.e., colloids.

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[1] Unpublished document WHO/Mal/Inform/32, March 1958

TABLE 9. SORPTION AND SPECIFIC SURFACE AREAS OF BUILDING MUDS SPRAYED WITH 2.14 G PER M² TECHNICAL DDT

Mud (sub-60-millimicron fraction)	CCl_4 sorptive capacity (%)	Hours to 100% knock-down of *A. aegypti* ♀ 4 weeks after spraying	Time of flow for 5 ml CCl_4 (seconds)	Surface area (Argungu= 1.00)	Persistency of DDT $\left(\frac{1}{\text{knock-down} \times 100}\right)$
Argungu	10.8	2.8	390	1.00	36
Birnin Kebbi	15.6	3.4	540	1.18	29
Yaba	24.5	4.0	960	1.58	25

TABLE 10. SORPTION AND SURFACE AREA OF BUILDING MUDS ESTIMATED BY METHYLENE BLUE SORPTION

Mud	Concentration of methylene blue * (mg/100 ml)	Surface area (m^2/g)
Argungu	15.5	15.5
Birnin Kebbi	14.5	14.5
Yaba	26.5	26.5

* Concentration at which the rate of sorption (which is initially the same for all muds) changes.

However, Langbridge[1] noted that certain muds with as high a specific surface area as 64 m^2/g exhibited a lower sorptive capacity than other muds with a much lower specific surface area (see page 877, 3rd paragraph).

It has been thought that measurement of the amount of colloids present in a mud should give enough indication of its sorptive capacity; however, it would appear that this is not so. Initially, sorption of residual insecticides is essentially a surface phenomenon involving the particles of mud on the surface and the immediate sub-surface. While the average proportion of the colloidal constituents present in a mud may give an indication as to the sorptive properties of the average type of such a mud, it does not indicate the amount of colloids in fact present at the surface.

Determination of persistency and sorptive capacity

The progressive elucidation of the mechanism of sorption and of its practical implications shows the need for suitable methods to determine either the sorptive capacity of the mud or the persistency of the insecticide or both.

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[1] Langbridge, D. M. (1958) In: Nigeria, Federal Malaria Service, *Western Sokoto Malaria Control Pilot Project, Insecticide Chemistry Laboratory, Annual report 1957-1958,* Yaba-Lagos (mimeographed document)

In most malaria campaigns such methods, by making it possible to plot a dosage/mortality/time curve, would be useful for a better planning of types, dosages and cycles of insecticide.

Sorptive capacity. In 1955, Barlow & Hadaway developed an empirical test to determine the sorptive capacity of a mud. The principle was that the amount of a non-polar compound, such as carbon tetrachloride, which can be taken up at the saturated vapour pressure at a given temperature is indicative of the capacities of sorption for non-polar compounds such as chlorinated insecticides. Using this test, Langbridge [1] found a fairly close relationship between sorptive capacity, specific surface area and rate of disappearance (see Table 7). But Barlow & Hadaway noted that with poorly active soils the test is of limited value, although it can be relied upon for highly active soils.

Langbridge [2] has attempted to measure sorptive capacity through the sorption of methylene blue from aqueous solutions, using as an index the surface area in m^2/g (see Table 9). He points out that this method will give a measure only of the total area accessible to the methylene blue molecules, which, being of a large size, are unable to penetrate the finer pores. The method will therefore register only a certain proportion of the internal surface.

A new line of investigation is being followed by Miles & Pearce (1957) by applying radioactivity to the study of sorption. The loss of insecticide from the surface is shown by the loss of radioactivity of mud surfaces treated with ^{14}C-labelled DDT, the partial shielding effect of the mud layers for weak beta-rays being measured. The method would seem to commend itself by its simplicity and rapidity, and the preliminary results are encouraging; but it is still in the trial stage.

Thus each of the techniques envisaged has its own inherent limitations and cannot be relied upon without serious reservations. All these techniques are still in the development stage. Moreover, the significant variations in persistency noted even within narrow limits, in places very close to each other, show the risk involved in attributing too much importance to such tests; strictly speaking, their results are valid only for the muds examined and for the specific small area where the samples of muds have been taken.

Determination of persistency. All the techniques and methods which have been devised for surface sampling of insecticide deposits or for determination of their persistency were originally devised for laboratory investigations, and only later were attempts made to adapt them for the field. The following techniques are used.

Visual and microscopical examination of sprayed surface (suitable when spraying with pure crystals, e.g., *pp′*-DDT). Microscopical examination of a sprayed surface is of value

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[1] Unpublished document WHO/Mal/Inform/32, March 1958

[2] In: Nigeria, Federal Malaria Service, *Western Sokoto Malaria Control Pilot Project, Insecticide Chemistry Laboratory, Annual report 1957-1958,* Yaba-Lagos (mimeographed document)

for laboratory investigations only, since inert diluents and fillers present in water-dispersible powders have a masking effect on crystals.

Adhesive impressions (applicable for DDT, dieldrin, BHC freshly sprayed only). The peel-off techniques using adhesives (transparent adhesive tape, silicone-parchment paper, or carboxy-cellulose or any other gum) have been the subject of careful investigations by Hadaway & Barlow and Alessandrini & Langbridge (unpublished reports). Their common and major inconvenience even in the laboratory is that the amount of insecticide deposit removed from the surface is influenced by the pressure exerted by the operator and by other factors such as the thickness of the layer of adhesive, and its possible change in adhesiveness with time, temperature, relative humidity, etc. Comparative trials in the laboratory reveal frequent inconsistencies. Even with pre-prepared surfaces in the field the irregularity of surfaces makes them unsuitable. For these reasons these techniques have not been recommended for use in surface sampling.

Scraping of the surface (suitable for DDT, dieldrin, BHC). Scraping removes the insecticide on the surface but it also removes a variable amount of insecticide present below the surface.

It can thus be seen that none of the available techniques for surface sampling of insecticides with a view to their chemical determination is reliable for assessing the amount of insecticide deposit on the surface. This is also the conclusion reached by an advisory group to WHO, which met in 1957 to consider the problem of sorption on mud walls.[1]

This conclusion should not be interpreted as signifying that the only amount of insecticide that matters is what is present on the surface—that which may be picked up by alighting mosquitos. The question raised here concerns the mode of contact of mosquitos with the insecticide (mechanical contact by pick-up or fumigant contact). It is accepted that DDT exerts its action mostly by mechanical contact and pick-up; for DDT, therefore, the amount *on* the surface is for practical purposes the amount which must be known. For dieldrin there is a suggestion that a fumigant effect plays a part in the lethal action of this insecticide;[2] and for BHC, the vapour effect is essential. The fumigant effect, depending on the vapour pressure, will take place whether the insecticide is on the surface or below, and whether it is completely or partly sorbed or totally unsorbed. This applies not only to chlorinated insecticides but even more strongly to organo-phosphorus insecticides, whose vapour pressures are generally even higher than that of BHC. Consequently, the amount active is not only that *on* the surface but also that *below* the surface.

On sorptive surfaces, the amounts of insecticide present on and below the surface are in a dynamic equilibrium (sorption-desorption). The process is governed by the competitive affinities of water and insecticide for the mud surface, and it is influenced by temperature and relative humidity. Much evidence on the phenomenon has been brought forward by Hadaway and Barlow in their various studies. Thus, the amount of insecticide below the

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[1] Unpublished document WHO/AS/156-57, October 1957

[2] Langbridge, D. M. (1958) In: Nigeria, Federal Malaria Service, *Western Sokoto Malaria Control Pilot Project, Insecticide Chemistry Laboratory, Annual report 1957-58*, Yaba-Lagos (mimeographed document)

surface is (essentially for BHC, to a lesser extent for dieldrin, and apparently still less for DDT) of importance. Not only is it important upon the first application, it is also and even more important upon further applications. When the first layers of mud have received successive dosages of insecticide the concentration of insecticide may increase temporarily to the point of saturation. At this stage, while the process of further penetration within the wall will continue, the speed of penetration of the insecticide molecules present on the surface is slowed down. In other words, the amount of insecticide potentially and biologically effective is related to the total amount of insecticide present on and below the surface.

These remarks show that it is necessary to reconsider the problem of sampling of insecticide deposits and to study from a different angle the relationship between dosage mortality and time. It is suggested that the estimation of the insecticide present on sorptive surfaces be made from blocks of mud taken from the sprayed wall down to a minimum depth that will ensure that the total amount of insecticide actually present will be determined.

Increasing the Persistency of Insecticides

In most tropical and subtropical areas many houses or huts have sorptive surfaces of mud wall, but quite frequently they are made of mixed materials, and generally, even when the wall is of mud, the roof may be of wood, thatch, banana-leaf, bamboo, grass, or some similar substance whose surface is non-sorptive. Sorption being rather an advantage for BHC, this insecticide might in principle be given preference on sorptive surfaces, but surfaces are rarely uniform in their degree of perviousness.

It would be unpractical, not to say impossible, to apply BHC on sorbent surfaces and DDT or dieldrin on impervious ones. Thus from the operational viewpoint the ideal insecticide is one which would combine the fumigant properties of BHC on sorptive surfaces and the prolonged contact action of DDT and dieldrin on non-sorptive ones. *Prima facie*, a way out of the difficulty could be found in the use of mixtures of BHC with either DDT or dieldrin. But development of resistance has limited freedom of action in this respect, while the technical, practical and economic implications of mixtures must not be disregarded. Under these circumstances a reasonable policy would be to take advantage of the maximum effect of each individual insecticide, by preventing sorption of DDT and dieldrin and by increasing the persistency of BHC on impervious surfaces.

The persistency, and consequently the effectiveness, of DDT and dieldrin being mostly related to the amount of the toxicant present on the surface and available on mechanical contact, any measure which could prevent sorption or slow down the disappearance of these insecticides would increase their effectiveness. This aim may be achieved either by influencing the surface or the formulation (both ways have been tried) or by applying high dosages in one application or repeated smaller dosages.

Prevention of sorption by action on the surface

Even before much was known regarding the mechanism of sorption several attempts were made to modify the surface to be sprayed in order to make it non-sorptive. *A priori*, however, it is clear that both practical and economic considerations militate against this principle since it involves additional and expensive manpower. It will also be easily seen that there is no simple means to render surfaces non-sorptive. A certain number of the means which have been tried for preventing or delaying sorption are, however, mentioned for the sake of completeness.

Blocking of the sorbent wall with a colloidal material has been tried in two ways. First, a 2% solution of size has been applied on the mud wall and left to dry for four days. Sorption was delayed, but the availability of the insecticide particles to the mosquito was reduced (Barlow & Hadaway, 1955). Secondly, trials have been carried out with urea-formaldehyde (50%) alkyd (50%) resins, containing 10% DDT or 5% dieldrin, applied on mud walls. Sorption was delayed only (Barlow & Hadaway, 1955).

Keeping the surface wet by treating the mud wall surface with a 10% solution on $CaCl_2$, which is a deliquescent solid, has had no effect on sorption, as is understandable in the light of Barlow & Hadaway's (1955) explanations of the influence of relative humidity.

Making use of assumed chemical degradation (for DDT) owing to the presence of iron oxides—a technique implying a chemical reaction between certain complex-forming and precipitate-forming reagents for ferric ions—has also been envisaged. To this end solutions of 10% potassium ferrocyanide, 10% sodium dimethyldithiocarbamate and molar dipotassium hydrogen phosphate were tried, but with no success.

Limewashing of walls appears to be so far the best practical means, in the absence of non-sorbable formulations, to delay sorption. Bordas et al. (1953) have used it with some success, and Barlow & Hadaway (1955) give some credit to this means. Enhanced persistency of dieldrin deposits has been noticed by Langbridge on a wall having a thick layer of whitewash (3-4 mm). The deposit found 16 weeks after the original application was 50% greater than the average on ordinary surfaces; but no enhancement was found on the thin coating of whitewash generally used by the local population in Northern Nigeria.[1]

Prevention of sorption by action on the formulation: resin-dieldrin wettable powders

Increased knowledge of the fundamental mechanism of sorption, of the intricate interactions between the mud surface and the insecticide formulation, and of the difficulty of determining the sorptive capacity and persistency suggested that all efforts to prevent sorption should be concentrated on the development of insecticide formulations not or little subject to sorption. One of the means tried has been to melt various resins (Aroclor, gilsonite, colophony and others) and dieldrin to form a metastable composition, and then to grind the mixture with an inert diluent, the assumption being that

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[1] Langbridge, D. M. (1955) In: Nigeria, Federal Malaria Service, *Western Sokoto Malaria Control Pilot Project, Insecticide Chemistry Laboratory, Third quarterly report,* Yaba-Lagos (mimeographed document)

TABLE 11. PERCENTAGE KNOCK-DOWN AND 24-HOUR MORTALITY OF HOUSEFLIES ON SORPTIVE MUD PANELS TREATED WITH DIELDRIN/COUMARONE RESIN MELT

Product	Age of residue	Hours after exposure						
		1½	2	3	4	5	6	24
Standard dieldrin wettable powder (25 mg dieldrin/ square foot)	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 (12.5 mg dieldrin/ square foot)	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**
	4 months	4	27	67	87	—	100	100**
	7 months	0	2	—	52	57	82	100**
	10 months		0	13	28	44	56	100**

* Reproduced, by permission, from Gerolt (1957)
** Panels used for exposure more than once.

the resin may act as a barrier to the penetration of the insecticide. Various formulations have been prepared with different resin-dieldrin proportions. Laboratory trials have been extremely encouraging, as may be seen from Table 11. Rather disappointing results, however, have been obtained so far from limited field trials in Tanganyika and Mexico inasmuch as the performance of the resin-dieldrin formulation was not better than that of the standard dieldrin water-dispersible powder. An experimental trial with a newly developed resin-dieldrin formulation is in progress in Zanzibar and more information will be forthcoming in the near future.

Increasing the persistency of BHC on impervious surfaces

The principle here is the same as for dieldrin-resin formulations and consists in mixing gamma-BHC with resins (Aroclor, Cereclor, etc.). The chemical condition of the insecticide is again unchanged and thus its vapour pressure is not modified, but in this case particular advantage is derived from the reduced release of vapour owing to physical barrier action. Successful trials have been made by addition of various polyphenyls (Hornstein

& Sullivan, 1954) and good results obtained in the laboratory. A 25% gamma-BHC-resin water-dispersible powder has recently been developed and is at present under trial in Iran.

Dosages, superimposition and persistency

Barlow & Hadaway (1955) have shown that sorption of DDT proceeds from layer to layer and have recovered DDT at a depth of 1.2 cm some 12 months after application. Recently, Langbridge (communication to WHO, 1958) has confirmed this again; in four years a surface deposit of 2.0 g DDT/m^2 has gone through a wall 20 cm thick giving an outside deposit of 0.12 g/m^2 on the opposite side. There is no doubt that on active mud sorption is a continuous process, as anticipated on theoretical grounds.

Attempts have been made to see whether it was possible to effect saturation, or at least a certain degree of saturation, of top layers of mud to decrease the speed of sorption. Langbridge[1] studied in Northern Nigeria the distribution of DDT (at 2 g/m^2) and dieldrin (at 0.5 g/m^2) regularly at various depths in walls and after a certain number of applications.

TABLE 12. MG OF DDT PER SQUARE FOOT * WITHIN WALL SURFACES 26 WEEKS AFTER PREVIOUS APPLICATION (IN NIGERIA)

Depth of wall sampled (mm)	Number of six-monthly spraying cycle				
	2	3	4	5	6
Surface-0.4	80	210	—	340	425
0.5-0.8	80	110	—	250	235
0.9-1.2	50	60	—	155	180
1.3-1.6	—	50	—	95	—

* A convenient conversion rate is 100 mg per square foot = 1 g per square metrc.

TABLE 13. MG OF DIELDRIN PER SQUARE FOOT * WITHIN WALL SURFACE 23-24 WEEKS AFTER SPRAYING, AT COMPLETION OF SIXTH SPRAYING CYCLE (IN NIGERIA)

Depth of wall sampled (mm)	Sample number												
	1	2	3	4	5	6	7	8	9	10	11	12	Average
Surface -0.4	26.1	10.7	10.5	13.9	19.2	13.8	13.3	11.5	9.1	7.3	9.1	5.4	12.5
0.5-0.8	13.8	11.6	10.1	6.4	16.1	9.0	10.5	7.8	7.0	3.1	6.2	1.6	8.6

* A convenient conversion rate is 100 mg per square foot = 1 g per square metre.

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[1] Unpublished document WHO/Mal/Inform/32, March 1958

His results, given in Tables 12 and 13, showed that there was a substantial increase in the amount of DDT within the surface half-millimetre. (The results in these tables are expressed in mg of DDT or dieldrin per square foot, which may be converted for all practical purposes at the rate 100 mg per square foot = 1 g per square metre.)

Six months after the sixth application three times the initial dosage of technical DDT was in the surface half-millimetre, but only one-quarter of the dieldrin dosage. This indicates that at commonly used dosages it was impossible to saturate muds, although additional successive doses might achieve a partial degree of saturation of the first layers and therefore indirectly result in increased persistency. The question whether saturation can be effected by massive dosages has not been sufficiently investigated. Although obvious reasons of economy are against it, it might be necessary under some conditions to increase present dosages on highly sorptive muds. Thus, as Langbridge put it, " a better biological effect would always be expected after repeated applications simply because diffusion of insecticide into the insect is likely to depend upon the concentration of insecticide in the superficial layer of mud ".

Apparent Discrepancies between Laboratory and Field Results

Since sorption may be so rapid (under laboratory conditions on certain muds it may occur in as little as a few hours) and since in most instances it involves inactivation of DDT and dieldrin deposits, how, then, can the success of malaria campaigns with these insecticides even in areas where huts or houses are built with sorptive material be explained?

No simple reply can be given to this question. From the wealth of data, observations and well-established facts already accumulated, it would appear that this success is attributable mostly to differences between experimental laboratory conditions and field conditions; in other words, this apparent conflict is the reflexion of variations between the artificial and the natural environment and of the differences—sometimes considerable—in testing methods and in the physical composition of test materials.

It should be remembered that similarly conflicting results have been observed in comparing the laboratory and field performance of resin-dieldrin formulations; the highly promising laboratory mortalities were not met in experimental huts. Table 14 brings out some of the differences.

In short, the conflict lies not in the facts but in their interpretation, in the tendency to draw general conclusions from scanty data. Factually, it is true to say that in the laboratory sorption has been shown to take place on certain muds at high speed with consequent inactivation of the insecticide deposit; under field conditions no evidence has yet been adduced to show that sorption is for all practical purposes either negligible or

TABLE 14. DIFFERENCES BETWEEN LABORATORY AND FIELD CONDITIONS

Conditions		Laboratory (artificial)	Field (natural)
1. Physical:	Temperature Relative humidity	Constant or variable (controlled stability or change).	Not under control (slow or rapid change).
	Ventilation	Controlled.	Not under control.
	Climatic condition (with time)	Controlled = no weathering effects.	Uncontrolled climatic conditions; weathering effects.
2. Test material		Artificial mud blocks (= pure, uniform mud by sieving and grinding, etc.) or original material.	Original material (= mud, sand, gravel, stones, straw, grass, wood, cow-dung, etc.)
3. Testing methods		Bio-assays or mosquito release in standard conditions and forced contact. Frequently performed in parallel with chemical analysis of deposits.	Bio-assays or mosquito release and forced contact (conditions not standard). Natural conditions and bionomics of the mosquito decisive. No chemical analysis.
4. Insecticide:	Form	Frequently technical product or pure isomers. Crystals: frequently mean proportion in optimum biological size (0-20 microns).	Water-dispersible powder formulation = crystals (from small to large size = less uniformity) plus inert diluents.
	Dosage	Accurately applied and checked.	Large variations and frequently unchecked.
5. Environment: human and animal influence		Practically none.	Smoke deposits, dust, rubbing off, etc.

important. It can, however, legitimately be assumed that the phenomenon does in fact take place in the field, and consequently, until there is evidence to the contrary, that it may create a problem in malaria eradication campaigns. The problem is one of the reduced effectiveness of the insecticide deposit and of the corresponding economic loss. It would certainly seem serious enough in its implications to warrant full consideration.

Dosages and Cycles

The question of sorption leads directly to the difficult problem of dosages and cycles. Although this cannot be fully considered here, it should be pointed out that there are wide variations between different countries in the dosages and spraying cycles used, and considerable difference of opinion exists as to which dosages or cycles are the best. It is also striking to note the scarcity of data in support of the established dosages, most of which are based on empirical observations and on tradition rather than on systematic observations.

A clearer appreciation of the need to consider sorption in connexion with dosages may be had by relating the daily mosquito mortality to the control of transmission. In practice, the insecticide has two effects: it reduces the density of mosquitos and it reduces their mean life expectation long enough for sporozoites not to develop. These effects depend on the potency of the insecticide and on the amount of it available in a lethal condition to the mosquito. But this amount is influenced by sorption and to a variable but essential extent by the bionomics of the mosquito; in other words, the critical values of anopheline mortality in areas where sorption is present can be determined only after an estimation of the factual influence of sorption on the effectiveness of the insecticide. " The critical survival rate of *A. gambiae* in an intensely malarious part of Africa is estimated at 0.62. In that region it is apparently wholly anthropophilic and rests very commonly indoors. The equivalent critical daily survival rate if it bit man at only every second feed would be 0.75, or at every fourth feed 0.82, but the mortality necessary, on such occasions as it entered houses, to secure these average values would be considerably higher, because exposure to risk would be less frequent. In this way the mild demand for an everyday 38% mortality would step up to a more severe one and insecticidal control would be put to its test." (Macdonald, 1957) It is also understandable that any reduction in the amount of insecticide available on each entry should increase the critical daily percentage mortality needed to interrupt transmission.

Effectiveness and Bio-assays

The fundamental investigations carried out so far on sorption have progressively elucidated the nature of the phenomenon and its mechanism, particularized a number of factors influencing it, and shown that it is qualitatively identical and only quantitatively different under laboratory or field conditions. This difference, which is one of degree only, is the reflection of obvious differences in conditions of experiment, of material and of assessment. They have also shown that while sorption is a major drawback for DDT and dieldrin, it is rather an advantage for BHC, and that the increase in relative humidity has a compensatory effect of varying degree.

The extent to which sorption influences the effectiveness of residual insecticides in malaria campaigns is not known; the problem of assessment of effectiveness is complex.

We have been dealing so far essentially with persistency, i.e., with those physico-chemical factors which influence the biological performance of the insecticide. With effectiveness another group of factors comes into play which are connected with the insect-insecticide response, that is, with the specific toxicity of the insecticide and its mode of action, the level of tolerance by the mosquito, and the bionomics of the mosquito. These may be briefly mentioned in conclusion.

Strictly speaking, effectiveness is measured by the mortality produced over a given period of time by a given dose of insecticide. In the extended and practical sense, effectiveness is, as previously said, the capacity of a given insecticide to control mosquitos to a level which renders transmission impossible. The picture of effectiveness is thus provided by entomological and epidemiological indices; this suggests why the correct evaluation of effectiveness is so complex a task, and helps to explain how difficult it may be to ensure adequate investigational conditions for the purpose.

Assessment of the effectiveness of an insecticide is an inherently difficult task, and attention has quite properly been devoted to developing relatively simple tools for the purpose. Thus it has been thought that mortality from bio-assay might serve as a useful index of effectiveness, and the wall bio-assay test has been recommended by the WHO Expert Committee on Insecticides " as a practical tool inasmuch as it supplements the chemical methods when they are of limited value " (World Health Organization, 1958). In other words, assessment of effectiveness is to be tried on the basis of the definable relationship between dosage and mortality under local conditions.

At this stage sufficient data still have to be accumulated to permit the expression of a scientific opinion on the value of this approach, it being clearly understood that such data are only one element in the total epidemiological picture which gives them their full significance.

Since this recommendation of the Expert Committee on Insecticides was made, a fairly large number of bio-assay tests have been carried out. It is still too early to draw any definite conclusions on the validity of the test itself or even on the meaning of the results attained. However, these preliminary data do show (*a*) an extreme variability of results obtained on the same wall by the same investigator (up to 40% variations in mortalities are frequent with a range of from 10% to 100%), and (*b*) the impossibility of correlating results with amounts of insecticide present, since no chemical assessment of the dosage actually applied or actually present has been made.

As to (*a*) it may be justifiable to think that the variability is only a consequence of the numerous variables involved in the test itself and in its application—i.e., the stage and reaction of mosquitos, the conditions of temperature and relative humidity, the minute local differences in the structure and nature of the surface, the variations in dosages of insecticide, and variations in the way the test is performed owing to variations inherent in the operator himself.

From (*b*) it follows that if the amount of insecticide actually present is ignored there is no way of correlating mortalities with dosages on a statistical basis and it is consequently impossible to plot any curve of the activity of the insecticide which would be of some general significance for the area under test.

From these remarks it is clear that any attempt to determine the number and importance of the variables involved must take account of the amount of insecticide present both at the very beginning and at the time of bio-assaying.

Conclusions

Considering the problem of sorption broadly in the light of this brief review, a number of conclusions may be drawn:

(*a*) The fundamental process and its dynamics are known: the physico-chemical rules of adsorption between solid and gaseous states are followed in sorption.

(*b*) The speed of sorption under laboratory conditions may vary from a few hours to a few weeks, depending on the mud and the insecticide.

(*c*) Sorption takes place in the field to a variable extent depending on very varied local conditions.

(*d*) Attempts made so far to prevent sorption by action on the surface or on the formulation have failed in practice.

(*e*) For lack of adequate investigations very little is known about the practical repercussions of sorption on malaria campaigns.

(*f*) Assessment of the practical repercussions of sorption is a complex task involving rather elaborate epidemiological and entomological investigations. Recourse to bio-assays of wall deposits has consequently been recommended with a view to providing a possible index of the residual activity of insecticides. Preliminary results have shown extreme variations suggesting that there may be a number of inherent unknown factors at play whose influence on the results requires analysis.

It is consequently suggested that mortality from bio-assay be assessed against the actual amount of insecticide present and chemically determined. Consistency of results from parallel chemical and biological tests should permit a curve of inactivation of insecticides to be plotted which would have statistical and thus general significance. Only then can a reasonable attempt be made to interpret the results or to use them as an index within the general epidemiological picture.

As yet the task is still one of assessment of the bio-assay technique through the accumulation of data and observations. To perform such a task, however, it would seem that the methodological approach in the field should be substantially the same as that in the laboratory.

RÉSUMÉ

L'étude systématique au laboratoire du phénomène de sorption des insecticides à action rémanente a permis de discerner les facteurs qui le conditionnent et d'en définir le mécanisme. La persistance des insecticides dépend de la nature de l'insecticide et de la surface sur laquelle il est appliqué. Elle est en outre sous l'influence directe de la température et de l'humidité relative. L'étude des caractères physico-chimiques de l'insecticide et des facteurs de surface montre que le phénomène est réglé par des lois bien définies et qu'il devrait donc être prévisible et mesurable.

Sur le terrain toutefois, la difficulté pratique d'y réussir est très grande en raison du nombre de variables qui s'y rencontrent et du manque d'une technique appropriée. En effet, la persistance de l'insecticide qui est un fait physico-chimique a pour répondant l'efficacité qui est un fait biologique. Les deux notions sont dégagées et l'auteur est ainsi amené à définir la signification de la méthode d'estimation de l'efficacité par essai biologique. Or, la détermination de la persistance s'est fondée jusqu'à présent sur la détermination de la quantité d'insecticide présent *sur* la surface à l'exclusion de celle qui est présente *sous* la surface. L'auteur expose les arguments favorables à une conception de la persistance fondée sur la détermination de la quantité d'insecticide totale présente *sur* et *sous* la surface. Les résultats très variables obtenus jusqu'à présent avec les essais biologiques suggèrent que la technique d'essai biologique elle-même demande encore à être standardisée. L'auteur insiste sur la nécessité d'une étude parallèle entre la persistance, c'est-à-dire entre la dose totale d'insecticide présent et l'efficacité, c'est-à-dire la mortalité résultant de cette dose. Au stade actuel, il est encore impossible par conséquent de donner une interprétation et donc une signification aux résultats des tests biologiques. L'auteur précise que les résultats de ces tests ne peuvent être compris que comme un indice additionnel dans l'ensemble du tableau entomologique et épidémiologique du paludisme dans une zone donnée.

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