THE LAWS OF DEPOSITION AND THE EFFECTIVENESS OF INSECTICIDAL AEROSOLS[1]

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This paper represents a review of studies made during 1943, 1944, and 1945 by the Central Aerosol Laboratories at Columbia University under NDRC contract and several cooperating agencies, including the U. S. Department of Agriculture, the Army, the Navy, the British Colonial office, and others.

The studies include several steps from fundamental research to field application and illustrate the spectacular results that can be obtained by the application of fundamental principles in this field, which had relied so much on empirical information.

The precision of the work here reported and the reliability of the results were made possible largely by the development by Sinclair and La Mer (13) of a laboratory generator which produced aerosols very uniform in particle size. For these experiments modifications were made in that generator to replace the spark source of nuclei, which produced toxic oxides of nitrogen, to make the output constant with time, and to extend the range of particle diameter up to 40 microns.

Figure 1 shows the homogeneous aerosol generator as used in these laboratory experiments. The nuclei were provided by heating a nichrome wire coil in which there was a plug of sodium chloride to a temperature just below red heat. A constant-voltage transformer was required to maintain constancy in the supply of nuclei for condensation.

The primary adjustment of particle size is obtained by adjustment of the heater temperature, higher temperatures resulting in larger particles. Necessarily, therefore, with increased particle size there was also an increased mass concentration. A bimetallic thermostat controlled the temperature of the box surrounding the first heater to ±0.5°C. and the flask to a much smaller tolerance.

The temperature of the second heater is not critical but it must be higher than that of the first to prevent undue condensation and must not be high enough to result in excessive decomposition. In the case of DDT in lubricating oil, this must not exceed 230°C. The output of the generator is assessed by passing its efflux through a glass wool filter in a drying tube and either weighing the filter or determining the DDT on the filter by chemical means.

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MORTALITY OF MOSQUITOES IN CONFINED ATMOSPHERES

In the first experiments (January, 1944) with mosquitoes (10) it was desired to test the effect of particle size on the toxicity of DDT aerosols to *Aedes aegypti* mosquitoes under conditions where there would be no relative motion of aerosol and insects. A homogeneous aerosol was confined in a wooden box of 85-l. capacity having glass windows. The aerosol was stirred first with a small fan at slow speed and then the cage of mosquitoes was introduced. A beam of light passing through the chamber onto a photronic cell indicated the decrease in aerosol concentration that might occur during the course of an exposure.

For each particle size several exposures were made at different values of time t and also of concentration C. The logarithm of the product Ct to kill 50 per cent of the female insects, log Ct_{50}, is related over a considerable particle size

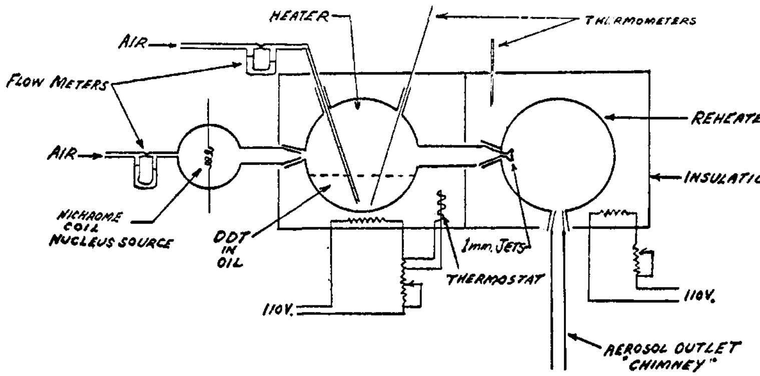

FIG. 1. Homogeneous aerosol generator

range to the particle diameter by the expression:

$$\log Ct_{50} = -2 \log d + \text{Constant} \quad (1)$$

Log Ct is plotted against log r [$\log r = \log d + \log 2$] in figure 2. The curve has a slope of -2, supporting the hypothesis that deposition on the insect by settling of the particles according to Stokes' law was in operation. The leveling off of the curve was believed due to the statistical effect, which would become evident at such a particle radius that a very small number of particles would be sufficient to kill an insect. A median lethal dose can be calculated then, based solely on gravity fall, assuming a cross-sectional area of 0.03 cm.^{2} for a mosquito as 3.6×10^{-6} mg. of 8 per cent DDT, or 2.9×10^{-7} mg. of DDT. This is $\frac{1}{100}$ the quantity believed on the basis of subsequent experiments to be more reliable.

The conclusions drawn from the data were that particle size should be about 10 microns diameter for optimum results. A large-scale aerosol generator was designed and built in accordance with this recommendation. The conclusion that the toxicity was proportional to the quantity depositing upon the insect was justified in spite of the fact that, as was learned soon thereafter, much of the aerosol probably reached the insects by way of convection currents and by way of their own motion rather than by gravity settling alone.

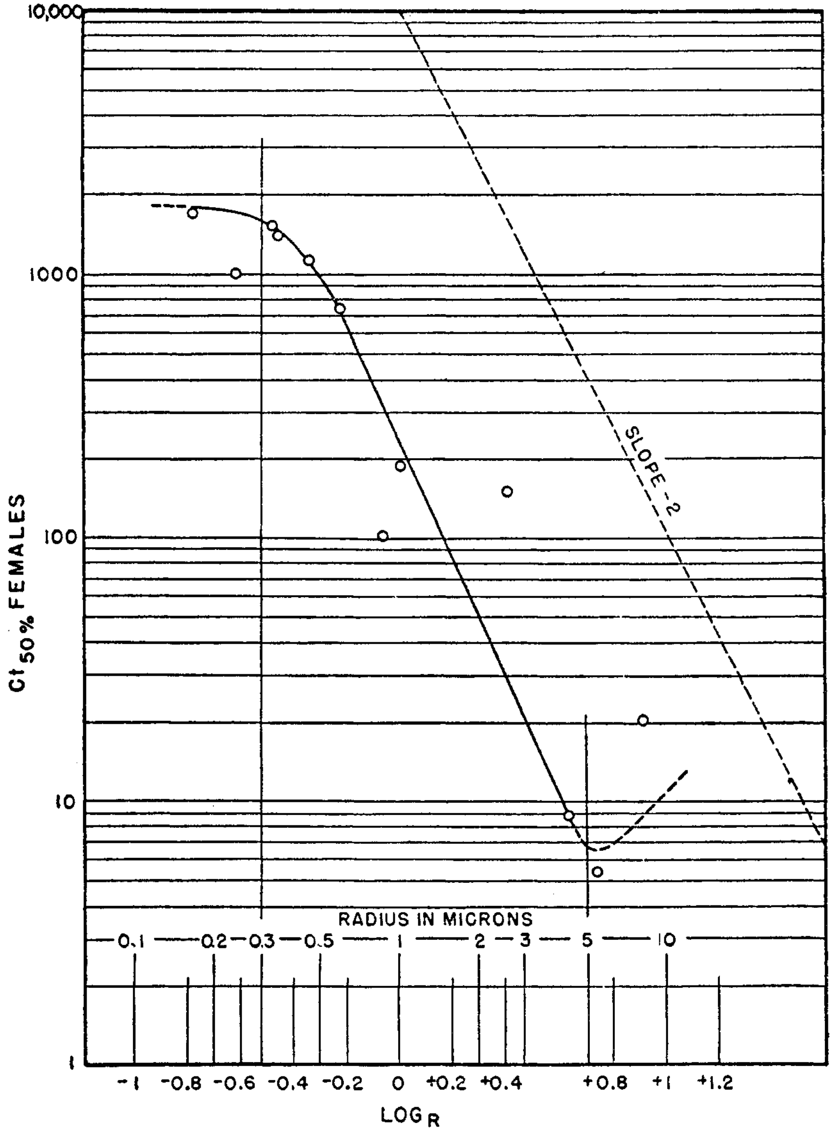

FIG. 2. Plot of log Ct against log r (La Mer *et al.*: J. Colloid Sci. **2**, 539 (1947))

THE MORTALITY OF MOSQUITOES AS A FUNCTION OF WIND VELOCITY AND PARTICLE SIZE

In order to test the effect of wind velocity and to obtain further data on the effect of particle size on the toxicity of DDT, more extensive experiments were performed in which mosquitoes in cages were exposed to aerosols within a wind tunnel 1 ft. square in cross-section (11).

Wind velocities of 2, 4, 8, and 16 miles per hour and particle sizes from 1 to 20 microns in diameter were used. For each particle size and wind velocity, enough exposures were made to determine the quantity of aerosol, M mg. per square foot of tunnel cross-section, required to kill 50 per cent of the female mosquitoes. This quantity was found to be determined by the product of the square of the diameter D and the wind velocity v (see figure 3).

The data obey the following law:

$$\log M = K \log D^2v + \text{Constant} \tag{2}$$

In the range $D^2v = 2$ to 200 microns² miles per hour, the data obey this relation with $K = -1$, M ranging between 300 mg. and 1 mg. per square foot. Between values of D^2v of 200 to 800 the value of K decreases to zero, and be-

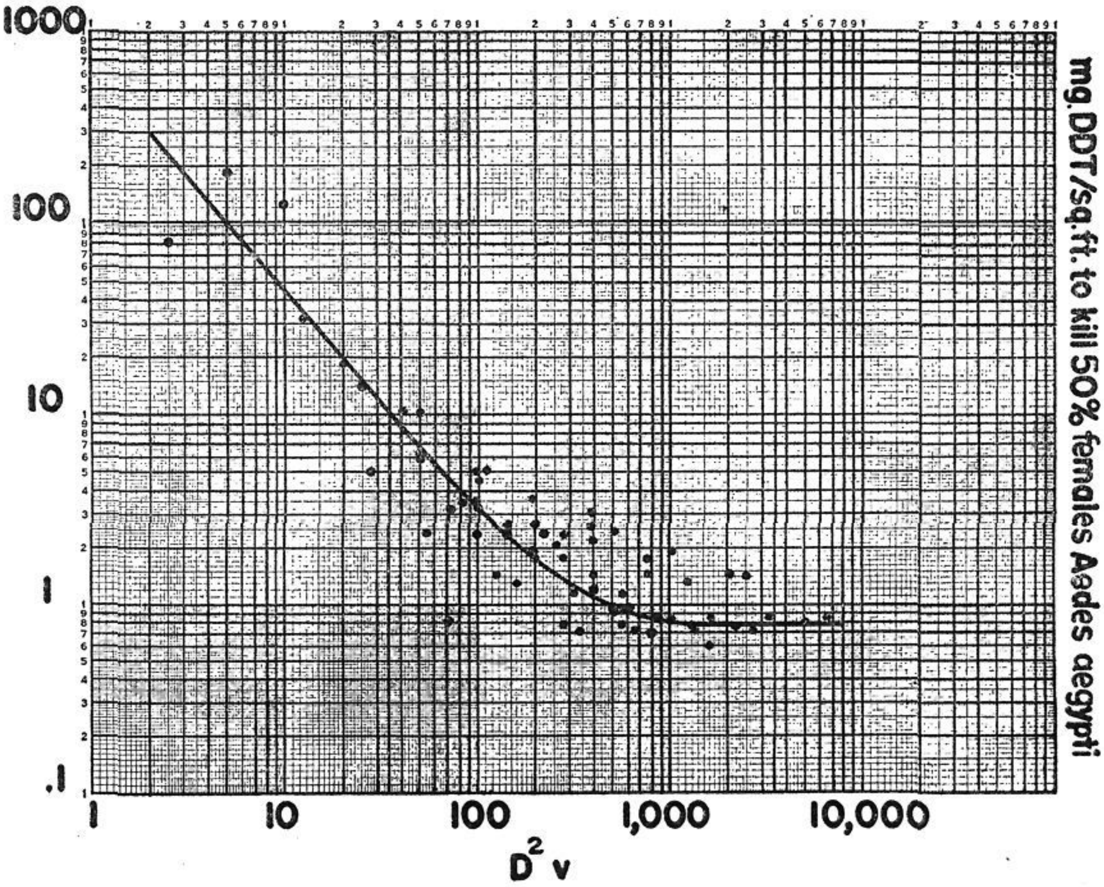

Fig. 3

tween values of 800 and 7000, K is zero, the value of M being 0.8 mg. of DDT per square foot.

That the spread of points about the curve of central tendency is due to the limited number of test insects is indicated by figure 4, where the mortality for males is plotted against that for females, each point representing a percentage mortality for a single cage containing roughly 50–100 insects. It is to be assumed that, had a sufficiently large number of insects been used, the data would have approached closer to the curve of figure 4 and also the curve of figure 3. Nevertheless, practically all the points lie within a factor of $\frac{1}{2}$ to 2 times the dose given by the curve drawn in figure 3. Use of mortality figures for males does not yield any better curve of the type of figure 3 than does the mortality curve for females, except that the former are more sensitive to DDT.

The results of these experiments were interpreted as follows: The motion of particles in an aerosol is governed principally by the motion of the air which supports them. This air, when a stationary object is in its path, tends to stream around the object and to carry the aerosol particle with it. The particle, on the other hand, being heavier than air and therefore having more inertia, tends to continue in a straight line. As a result, the path of the aerosol particle has a smaller curvature than the path of the air in which it is suspended. Therefore, when an aerosol is blown against an object a fraction of the aerosol strikes an object in its direct path, this fraction becoming larger as the velocity

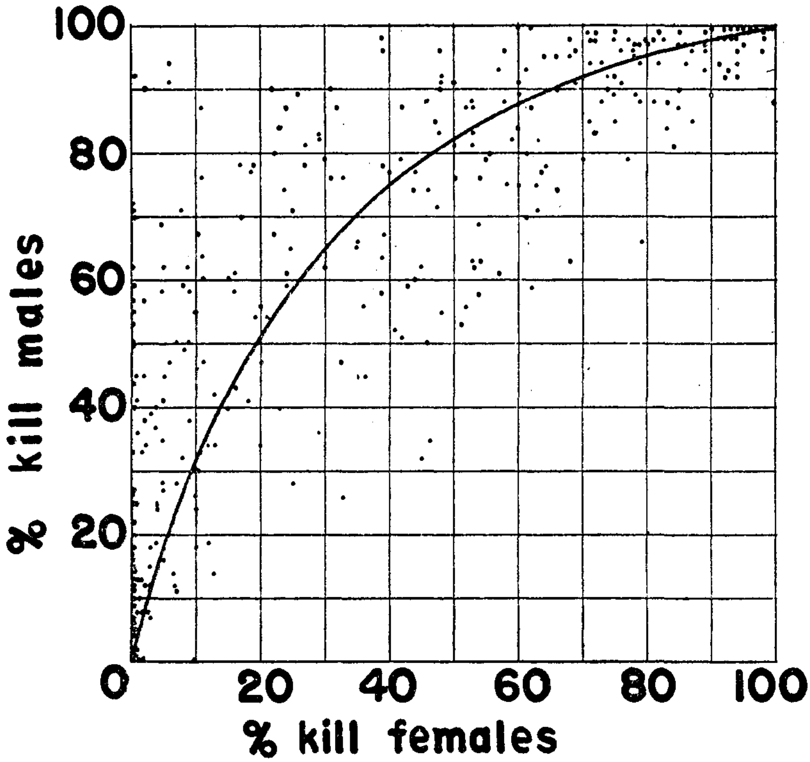

FIG. 4. Plot of mortality for males against that for females

of the aerosol is increased, as the particle diameter is increased, or as the size of the object is reduced. When this fraction approaches unity, further increase in particle size or wind velocity ceases to have effect.

A mathematical analysis which predicts this type of dependence of deposition upon D^2v has been given by Sell (12; see also 4 and 15) for simple geometrical objects.

The data on insects in the confined atmospheres must be interpreted as being complicated by a small velocity which resulted on account of (*a*) a small amount of flying, (*b*) putting the cages into the aerosol and taking them out, and (*c*) a certain amount of convection.

The following conclusions were drawn from the wind tunnel experiments:

1. The data indicate that the toxicity is determined by the quantity deposited on the insects which, according to Sell, would be expected in the limits of low and high values of D^2v to yield the slopes obtained in figure 3.

2. The data for larger particle size and wind speed were taken to indicate saturation, i.e., that the insects were taking up a quantity of aerosol equal to that which otherwise would have passed through their cross-section. The median lethal dose of DDT as yielded by this interpretation is 3.0×10^{-8} g. of DDT.

3. A wind tunnel could be used as a chamber for testing relative toxicity of insecticides in conjunction with an atomizer which would make particles within a certain range such that D^2v will be above 800 and yet the loss of particles by settling in the tunnel before the insect cages are reached would be negligible.

Experiments were performed in the wind tunnel using a specially made atomizer to test the toxicity of DDT in a variety of solvents (8).

It was found that DDT was most effective when it was present with a relatively non-volatile solvent, but that it lost effectiveness if used with solvents that would volatilize to relative completion before the aerosol contracted the insects. This type of toxicity determination is so convenient in operation and is so rapid that we have suggested that it be studied further as a substitute for the standard Peet-Grady chamber method.

4. In using aerosols for the kill of mosquitoes by contact with the aerosol, it is futile to use values of particle diameter such that D^2v would be greater than 700 microns^2 miles per hour. Larger values of D^2v would kill mosquitoes no better and would not have the ability to be carried into crevices and around objects as well. Under practical conditions this indicated a particle size of approximately 10 microns diameter.

GENERATION OF AEROSOLS FOR FIELD CONTROL

The laboratory data showed that it would be feasible to control insects over large areas by emission of a DDT aerosol from a generator moving crosswise to the wind and allowing the wind to carry it over long distances to the insects. However, particle size would have to be controlled carefully. A generator was developed (U. S. patent 2,416,256) that would produce particles in the most desirable particle-size range in outputs up to about 135 lb. of DDT per hour, or 50 gallons per hour of 35 per cent DDT. This machine pumped DDT dissolved in oil and water either separately or as an emulsion under pressure through heating coils where all of the water was converted into superheated steam and the mixture was ejected through a nozzle. The nozzle consisted of a tube about 2 in. long and $\frac{1}{8}$ in. in diameter. By adjustment of temperature and pressure, the average particle diameter could be adjusted from about 2 microns up to 60 microns.

The carrying power of the aerosol produced was measured by dissolving a dye in the oil, generating an aerosol containing the dye, and measuring the deposit of aerosol collected on suitable surfaces such as glass slides or plates at convenient distances downwind through various types of terrain and vegetation by colorimetric methods. Original data and plots similar to figure 5 are given in reference 1. Figure 5 gives the results of two of the typical experiments. It was found that in general the weight m per square foot deposited on a plate is an inverse exponential function of the distance x from the generator, so that

$$m = m_0 e^{-2.3kx} \tag{3}$$

where k is the slope of $\log m$ *vs.* x and m_0 is the deposit per unit area at very short distances ($x = 0$).

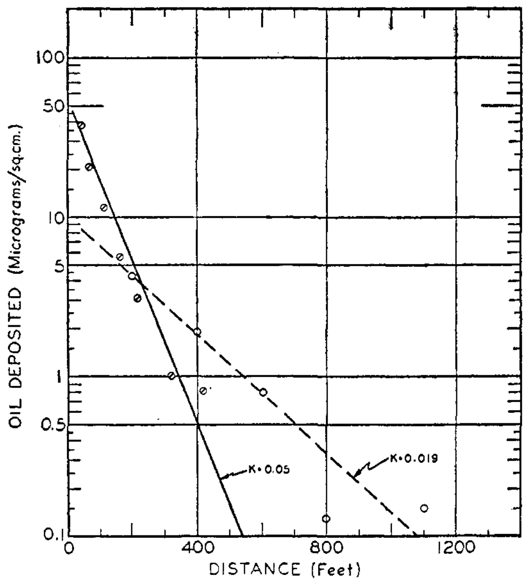

FIG. 5. Deposition as a function of distance. ○, test I (light forest); ⊘, test B (light-dense forest). Log $M = -Kd +$ Constant.

It is instructive to consider the simplified case where the ground surface is made up of surfaces similar to the plates used for the measurement and that loss occurs entirely on these surfaces.

Suppose that R is the output rate of a generator in pounds per foot of travel measured perpendicular to the direction of the wind, and m is the deposit rate in pounds per square foot.

The amount A lb. deposited in an area 1 ft. wide bounded by the generator travel line and a distance x is calculated by integrating $m\,dx$ between the limits zero and x giving:

$$A = \frac{m_0}{2.3k} - \frac{m_0}{2.3k} e^{-2.3kx} \tag{4}$$

If the distance x is taken as extremely large, then A becomes identical with R, the output rate per foot of travel, and

$$R = \frac{m_0}{2.3k} \tag{5}$$

Suppose it is desired to apply a dose of m_1 at a distance x from the generator. Then from equation 3

$$m_1 = m_0 e^{-2.3kx_1} \tag{6}$$

and from equation 5

$$m_1 = 2.3kRe^{-2.3x_1} \tag{7}$$

or

$$\frac{R}{m_1} = \frac{e^{2.3kx_1}}{2.3k} \tag{8}$$

Table 1 gives values for R/m_1 as a function of x_1 and k.

TABLE 1

Values of R/m_1 as a function of x_1 and k

x_1	R/m_1	
	$k=0.0001$	$k=0.005$
0	434	87
100	546	276
200	687	870
500	1370	27,600
1000	4340	8,700,000

TABLE 2

Most economical value of k to produce a given deposition rate at a distance x_1 together with required dosage

x_1	k	R/m
0		0
100	0.00434	272
200	0.00217	545
500	0.00087	1360
1000	0.000434	2720

Differentiation of equation 8 with respect to k yields an equation for the most advantageous value of k, k_{econ}, to yield lowest values of R for a given value of m at a distance x_1.

$$k_{econ} = 1/(2.3x_1); \qquad (R/M)_{econ} = e/2.3k_{econ} \tag{9}$$

Table 2 reveals that the output becomes proportional to the distance desired provided k is always optimum.

Comparison of output rates R between tables 1 and 2 shows that the value of k has a tremendous effect upon the amount of material required to kill insects at a long distance. Only through proper particle-size control can output values be kept sufficiently low if long-distance kill (over a few hundred feet) is desired.

FIELD TESTS

Field tests were performed under various conditions of terrain from open country to jungle and forested areas under various weather conditions to test these hypotheses (1, 2, 3, 5, 6, 7, 9, 14).

Selected tests as described below will indicate the power of controlled particle size.

I. The generator was transported for 500 ft. along the edge of a fringe of dense vegetation bordering a light forested area crosswise to the wind. Wind speed in the forest was 0.3 mile per hour or less. The dosage was estimated as 15 gallons of emulsion of 5 per cent DDT (total 5 lb.) for the 500-ft. front. Adult mosquitoes were trapped in cages and in barrels and were placed at seven positions from 100 to 1100 ft. at ground level and also at 6 ft. above ground level.

Pans of larvae were also placed at these positions. Table 3 gives the mortality figures obtained. A peculiar topographical condition was blamed for the relatively low deposition and poor kill at 800 ft. The dosages given in the table are those collected on a horizontal glass plate. The actual consumption of DDT can be assumed to be much greater, being absorbed on all types of surfaces (tree leaves, etc.), of which there can be many square feet per square foot of ground area. k in this case was 0.019 (see figure 5, test I).

TABLE 3

Forested area

DISTANCE	DDT	PER CENT KILL LARVAE	PER CENT KILL ADULTS	
			Barrels	Cages
feet	*lb./acre*			
100		100	100	100
200	0.038	100	100	100
400	0.017	100	100	100
600	0.0069	100	100	100
800	0.0011	100	190	60
1100	0.0013	95	100	85
1200				100

II. Another test was made in rather open marshy country covered with a low weed growth. The wind speed was 2 miles per hour, the generator path was 500 ft. long, and it emitted 0.5 lb. of DDT per 1000 ft. of front as an aerosol of 3–4 microns diameter. Counts were made of landing rates on a man's back over several previous mornings of 200–300 mosquitoes per minute. Stations were laid out at 1400, 1700, and 2400 ft. downwind from the generator. Complete elimination of free mosquitoes was indicated at the end of the run and all caged mosquitoes were dead or suffering visibly from poisoning 3 hr. after the test (see table 4). In addition, landing rates were recorded up to 5300 ft. Comparison of the figures with checks made upwind from the generator indicates that *100 per cent mortality was obtained for half a mile and at least 95 per cent mortality was obtained for an additional half mile.*

III. A set of two tests at Mantoloking, New Jersey, a resort town on the Atlantic shore, illustrates again the importance of particle size. Mantoloking is part of a sand spit extending north and south on the New Jersey coast and is separated from the mainland by Barnegat Bay. At Mantoloking the spit is 800 to 2000 ft. wide. In the first test, an aerosol of 10 microns diameter was used, emitted from the generator mounted on a truck. With an output rate of 34 lb. of DDT per mile, there was a complete elimination of adult mosquitoes in the area, but larvae in jars were hardly affected even at a distance of only 50 ft. In the second test, the generator was mounted on a skiff operating in the bay.

TABLE 4

Marshy country

DISTANCE	LANDING COUNTS		CAGED MOSQUITOES, PER CENT MORTALITY AFTER 3 HR.*
	Before test	3 hr. after test	
feet			
1400	35	0	90
1700	50	0	95
2400	112	0	90

* All living mosquitoes affected by DDT.

TABLE 5

Test B: Mantoloking

STATION NO.	34 LB. DDT PER MILE POSITION	PER CENT MORTALITY	
		Adults	Large larvae
1	Bay	70	100
2	Middle	100	100
3	Ocean	80	100
4	Bay	100	100
5	Middle	100	100
6	Ocean	100	100
7	Bay	100	96
8	Ocean	64	76

A steady wind blowing from the bay served to spread the aerosol over the land. Caged adults and larvae in jars were placed at eight positions, three on the ocean beach, on the far side of the spit, three at the shore of the bay nearest the path of the generator, and two others part of the way across the spit. Wind velocity was 3–5 miles per hour. Particle size for this test was 10–25 microns diameter. Table 5 shows the mortality figures.

Output was 34 lb. of DDT per mile, or the same as in the previous test. Larvae were killed this time even at the extreme distance, approximately 2000 ft. It is felt that doubling the quantity emitted without change of particle size would have provided 100 per cent control.

IV. Another experiment was conducted (3, 14) jointly with C. B. Symes and

L. D. Hadaway, the entomologists of the British Colonial Office in Georgetown, British Guiana. The aerosol generator was operated for a path of 1000 ft. on each of three streets which composed Lodge Village, a malaria-infested community on the outskirts of Georgetown. The area south of the village consisted first of 2000 ft. of cemetery, then 2000 ft. of rice fields, and then 4000 ft. of cane fields. Vegetation grew thickly to a height of 20 ft. around the houses. Rice fields were covered with vegetation about a foot high. The sugar cane was 6–8 ft. high. Sixty stations were laid out in this area up to 8000 ft. downwind from the generator. Extensive daily counts of 500 to 1400 mosquitoes per room were made in the native houses of the village. Complete kill of caged adults and almost complete kill of free adults up to 2300 ft. beyond the village was evident after the application of a total of 36.6 lb. of DDT. Up to 4000 ft. the kill diminished to 50 per cent and up to 8000 ft. went to zero. *Larvae were killed at all stations up to 8000 ft.* except in one most easterly station.

SUMMARY AND CONCLUSIONS

1. Data on the effect of particle diameter and wind velocity show that the toxicity of DDT aerosols to mosquitoes follows the law of deposition of aerosols on geometrical objects, as calculated by Sell.
2. A method of determination of toxicity is proposed making use of a wind tunnel instead of a Peet-Grady chamber. Problems of contamination can be largely avoided and rational results in terms of absolute toxicity obtained.
3. The deposition from an aerosol is an inverse exponential function of the distance from the source. The constant involved depends upon a number of factors, such as turbulence, particle size, density of vegetation, wind velocity, etc.
4. This exponential relationship makes necessary careful control of particle size as well as selection of favorable meteorological conditions if good results are to be achieved.
5. A generator was developed by which large-scale control of mosquitoes by ground dispersal is possible over difficult and inaccessible terrain. This was achieved through selection of particle size and by operation under favorable meteorological conditions.

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