82. P. R. Schwartz and A. H. Barrett, *ibid.* **157**, L109 (1969); J. A. Ball, C. A. Gottlieb, M. L. Meeks, H. E. Radford, *ibid.* **163**, L33 (1971).
83. A. C. Cheung, D. M. Rank, C. H. Townes, W. J. Welch, *Nature* **221**, 917 (1969).
84. A. C. Cheung, D. M. Rank, C. H. Townes, D. D. Thornton, W. J. Welch, *ibid.*, p. 626.
85. L. E. Snyder, D. Buhl, B. Zuckerman, P. Palmer, *Phys. Rev. Lett.* **22**, 679 (1969).
86. M. Kutner and P. Thaddeus, *Astrophys. J.* **168**, L67 (1971).
87. N. J. Evans II, A. C. Cheung, R. M. Sloanaker, *ibid.* **159**, L9 (1970).
88. W. J. Welch, *Bull. Amer. Astron. Soc.* **2**, 355 (1970).
89. M. Kutner, P. Thaddeus, K. B. Jefferts, A. A. Penzias, R. W. Wilson, *Astrophys. J.* **164**, L49 (1971).
90. B. Zuckerman, P. Palmer, L. E. Snyder, D. Buhl, *ibid.* **157**, L167 (1969).
91. R. W. Wilson, K. B. Jefferts, A. A. Penzias, *ibid.* **161**, L43 (1970).
92. K. B. Jefferts, A. A. Penzias, R. W. Wilson, *ibid.*, p. L87.
93. L. E. Snyder and D. Buhl, *ibid.* **163**, L47 (1971).
94. B. E. Turner, *ibid.*, p. L35.
95. J. A. Ball, C. Z. Gottlieb, A. E. Lilley, H. E. Radford, *ibid.* **162**, L203 (1970).
96. A. H. Barrett, P. R. Schwartz, J. W. Waters, *ibid.* **168**, L101 (1971).
97. B. Zuckerman, J. A. Ball, C. A. Gottlieb, *ibid.* **163**, L41 (1971).
98. A. A. Penzias, P. M. Solomon, R. W. Wilson, K. B. Jefferts, *ibid.* **168**, L53 (1971); K. B. Jefferts, paper delivered at the 17th International Astrophysics Symposium, Liège (1971).
99. R. W. Wilson, A. A. Penzias, K. B. Jefferts, M. Kutner, P. Thaddeus, *Astrophys. J.* **167**, L97 (1971).
100. K. B. Jefferts, A. A. Penzias, R. W. Wilson, P. M. Solomon, *ibid.* **168**, L111 (1971).
101. P. M. Solomon, K. B. Jefferts, A. A. Penzias, R. W. Wilson, *ibid.*, p. L107.
102. R. H. Rubin, G. W. Swenson, Jr., R. C. Benson, H. L. Tigelaar, W. H. Flygare, *ibid.* **169**, L39 (1971).
103. J. A. Ball, C. A. Gottlieb, A. E. Lilley, H. E. Radford, *Int. Astron. Union Circ. 2350* (1971).
104. P. Palmer and B. Zuckerman, *Astrophys. J.* **148**, 727 (1967).
105. B. Zuckerman, D. Buhl, P. Palmer, L. E. Snyder, *ibid.* **160**, 485 (1970).
106. P. Solomon, paper delivered at the 17th International Astrophysics Symposium, Liège (1971).
107. F. J. Kerr and R. Vallak, *Aust. J. Phys. Astrophys. Suppl.* **3**, 1 (1967).
108. S. H. Knowles, C. H. Mayer, A. C. Cheung, D. M. Rank, C. H. Townes, *Science* **163**, 1055 (1969).
109. B. Höglund and P. G. Mezger, *ibid.* **150**, 339 (1965).
110. The work discussed here was supported in part by NSF grant GP 24611, NASA grant NGL-05-003-272, ONR grant NOOO 14-69-A-0200-1047.

DDT in the Biosphere: Where Does It Go?

Global modeling permits an appraisal of the hazards of DDT residues in the biosphere.

George M. Woodwell, Paul P. Craig, and Horton A. Johnson

DDT has been used in large quantities as an insecticide since 1942. Its residues (*1*) are sufficiently persistent and mobile to have a worldwide distribution, appearing in the lipids of most organisms (*2–7*), in air (*7–9*), and occasionally in meltwaters of Antarctic snows (*10*). Concentrations in certain of the earth's biota have reached toxic levels, causing spectacular declines in populations of certain carnivorous and scavenging birds and fish, aggravating the problems of pollution, and threatening significant contamination of human food chains (*2, 4, 11–14*). Recognition of the seriousness of these problems has led to recent restrictions in the use of DDT in the United States and abroad. There is at least a possibility that most of the DDT that has been or will ever be produced has already been used and that little, if any, will be applied after the mid-1970's (*15*). The persistence of DDT residues is great enough, however, that the residues will continue to be redistributed for many years after use of the pesticide has stopped, presumably presenting a continuing hazard to all the biota. The extensive data available on the distribution and effects of DDT make it, together with radioactive substances, the best known of the biospheric pollutants and a valuable subject for a case history study (*12, 16, 17*). But basic questions remain, among them the following: What becomes of DDT released into the biosphere? How serious are the hazards? and, How long will the hazards persist?

In an effort to answer these questions we have attempted to develop a model of the circulation of DDT in the biosphere. We have done this on the basis of two limiting assumptions: (i) that use of DDT will decline to zero by 1974 and, alternatively, (ii) that, between now and then, use will increase throughout the world.

Certain physical properties of DDT are important in determining its behavior in the biosphere. First, because of the high solubility of DDT in fats together with its low solubility in water (*18, 19*), DDT residues tend to accumulate in lipids and therefore in plants and animals. Second, the residues are very persistent in nature: estimates of their half-life range upward to 20 years, perhaps longer under certain circumstances (*20–24*). Third, DDT has a vapor pressure high enough to assure direct losses from plants and soil into the atmosphere, which can carry residues worldwide (*12, 13*). Thus soils, air, the waters of the oceans, and the biota are all potential reservoirs for DDT residues, and the hazard to the biota, including man, hinges on the distribution of DDT residues among these reservoirs. How large are the reservoirs, and what are the rates of exchange between them?

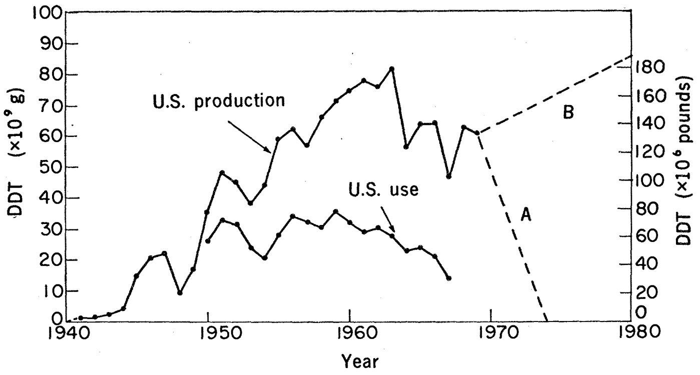
The answers are not available in any simple or absolute sense. Most are available, however, at least by inference. First, we must know how much DDT has been produced and something about its distribution.

Input: DDT Production

The amount of DDT produced in the United States each year is reported by the U.S. Tariff Commission (*25*). In the crop year 1963 the amount of DDT produced reached a maximum 8.13×10^{10} grams (179×10^6 pounds) (Fig. 1). Production has dropped in the United States since 1963, but more than 6.0×10^{10} grams of DDT were produced in 1969. Preliminary figures for 1970 reveal that DDT production declined by more than 50 percent. About 70 percent of the amount of DDT produced appears to have been used outside the United States. The total

---

The authors are members, respectively, of the departments of biology, physics, and medicine of Brookhaven National Laboratory, Upton, New York 11973. Dr. Craig is presently on leave to the National Science Foundation. Dr. Johnson is presently with the Health Sciences Center of the State University of New York at Stony Brook.



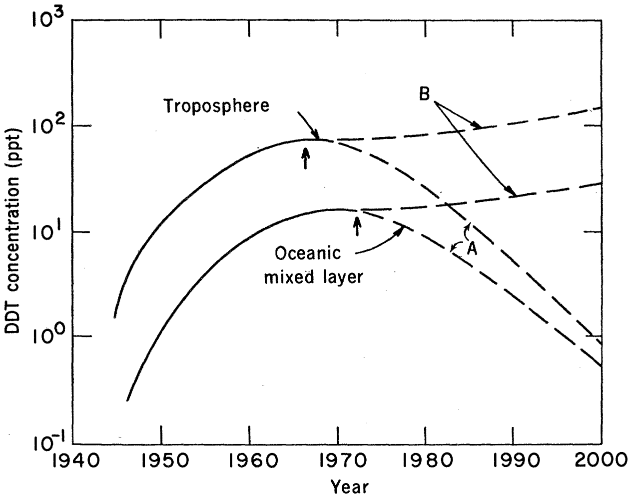


Fig. 1 (left). DDT production and use in the United States: curve A, based on the assumption of declining use through 1974; curve B, based on the assumption of increasing use through 1980. Dotted lines indicate projections. Fig. 2 (right). DDT concentrations in the mixed layer of the oceans, in the troposphere, and in the biota projected through the year 2000 on the basis of assumptions A and B of Fig. 1.

amount of DDT produced in the United States, integrated over the entire period through 1974, when we have assumed that DDT will no longer be used, is estimated to be 1.4×10^{12} grams. No data on world production are available. We have assumed that the amount of DDT produced in the world, including the U.S. fractions, is twice the amount produced in the United States, or about 2.8×10^{12} grams in total through 1974.

This DDT has been distributed widely around the world, most heavily in humid temperate and tropical zones, It is commonly applied by spraying a liquid suspension or solution from mobile ground equipment or from aircraft. The fraction of the spray that lands on the target varies, but some fraction of both aerial and ground applications remains airborne. Aerial applications of DDT to forests in the northeastern United States show that 50 percent or less of the amount emitted from the planes is deposited on the forest. The rest is dispersed into the air. Much of the airborne fraction returns to the ground nearby, but small droplets or particles are likely to remain aloft, to become associated with other particles, and may be carried great distances (*7–9, 14, 26*).

DDT in Soils

The persistence of DDT led to early recognition that residues might accumulate in soils. On the basis of a review of published data (Table 1), we have estimated that agricultural soils in the United States contain an average content of DDT approaching 0.168 gram per square meter (1.50 pounds per acre) (*22, 23, 27–35*). Nonagricultural soils were estimated to contain an average of 4.5×10^{-4} gram per square meter (*36*).

These estimates can be used to calculate the rates of loss of DDT residues from soils in the United States. Within the United States, the total contiguous land area is 7.7×10^{12} square meters (1.9×10^{9} acres), about 11 percent of which (0.85×10^{12} square meters) is kept in crops on which insecticides are used (*37*). The agricultural land apparently retains about 1.42 $\times$ 10^{11} grams of DDT. These data were obtained in the early 1960's when DDT production was high. The rate of use of DDT in the United States during this period was 2.7×10^{10} grams per year (6×10^{7} pounds per year) (Fig. 1) or 0.0318 gram per year for each square meter of agricultural cropland (0.28 pound per acre per year). The mean lifetime of DDT in soils must be about $10^{11}/(2.7 \times 10^{10}) = 5.3$ years. This estimate approximates Edwards' earlier estimate of 4.3 years (*21, 38*); it is substantially less than the estimated lifetime of 10 years for DDT in certain soils (*22, 24*). We have assumed a mean resident time of 4.5 years for DDT on land.

Four mechanisms probably account for most losses of DDT residues from soils: (i) volatilization (including losses by wind erosion of small particles from the soil surface), (ii) removal by harvest of organic matter, (iii) water runoff, and (iv) chemical (including biotic) degradation.

The occurrence of DDT residues in rainwater suggests that large amounts of DDT may move through the atmosphere either adsorbed to particles or as vapor. The small number of data available on the volatilization of DDT suggest a time constant for volatilization of several years, but the evidence supports the conclusion that vaporization is more important than such a long time constant would indicate (*14, 21, 38, 39*). The rates of disappearance of residues of dieldrin (*1*) by volatilization have been shown to be a function of the rate of air movement through soils (*40*). Residence times for DDT in organic soils where movement is slow are greater than residence times in mineral soils, a relationship that could only be the case if biotic degradation of DDT residues proceeds slowly in these soils in comparison with volatilization. Apparently evaporation is a major mechanism for the removal of residues of the persistent pesticides from soils (*21, 38*), and, despite its slowness, evaporation proceeds faster than chemical breakdown.

DDT residues are also removed from soils by the harvest of organic matter, but the evidence suggests that this is not a major route of transport. The net amount of crop harvested, even for highly productive agricultural crops, seldom exceeds 5000 grams per square meter and more often approaches 1000 grams per square meter (*41*). If we assume an annual harvest of 3000 grams per square meter containing 1 part per million (ppm) of DDT, currently the maximum concentration allowed in many foods, the harvest would remove 0.003 gram per square meter of DDT, about 1 percent of an annual application of 0.33 gram per square meter, or about 10 percent of the estimated annual average amount of DDT used for all crop land (see above). Thus, even a harvest of 100 percent of the primary production would remove only a small fraction of an annual application of DDT. Actual harvests that remove the organic matter from the site and might transport DDT residues to urban areas or to watercourses are much less efficient. We have assumed a removal of 1 percent of the total DDT used on the crop.

DDT is not normally applied to forage crops or in places where it can contaminate tissues of animals used for food, but farm animals do, nonetheless, become contaminated, as do most animals. The amount of farm animal biomass produced each year usually approximates 10 percent or less of the net primary production, and it is difficult to visualize under present levels and patterns of use a circumstance in which harvest of farm or range animals would account for more of the DDT residues than would the annual harvest of the plant crops, despite the possibility of concentration of the residues through food chain effects.

The transport of DDT residues from agricultural soils in surface waters has often been assumed to be of major importance in the accumulation of residues in the oceans. Heavy rains do remove DDT either adsorbed to soil particles or in solution but the amounts are small in comparison with the total amounts of DDT produced. Surface runoff over the entire United States is about one-third of the annual precipitation, or 23 centimeters (9 inches) per year out of an average precipitation estimated as 76 centimeters (*42*). The total volume of this water is 2×10^{12} cubic meters per year. If it were saturated with DDT residues, it would contain about 1 part per billion (ppb) (*18, 43*) and would remove 2×10^{9} grams of DDT per year in solution. DDT is applied only at certain times of the year in certain areas and seldom directly to bodies of water. Observed river water concentrations of pesticides (including particles) range from concentrations below the limit of detection [less than 10 parts per trillion (ppt)] to almost 100 ppt (*18, 43*). An average concentration of 50 ppt implies an annual runoff of about 10^{8} grams per year, which accounts for about 0.1 percent of the amount of DDT produced per year. Similar conclusions have been reached by Risebrough *et al.* (*9*), who also decided that movement of residues in the atmosphere is the most important transport route, and by the ocean pollution group of the M.I.T. Study of Critical Environmental Problems (*14*).

DDT in the Atmosphere

The vapor pressure of DDT at 20°C is 1.5×10^{-7} millimeter of mercury (*44*), producing an equilibrium concentration of DDT in the atmosphere of about 3×10^{-6} gram per cubic meter or about 2 ppb by weight. The vapor pressure drops with decreasing temperature. If we assume that DDT in the atmosphere remains as vapor, the saturation capacity of the atmosphere to the tropopause would be about 10^{12} grams of DDT, or about as much as has been produced to date. But DDT residues also exist in association with atmospheric particles, and the earth's atmosphere can probably contain very much larger quantities than the saturation capacity alone would indicate. This means that the atmosphere is potentially a large reservoir in addition to being a major means of transport for the residues.

Residues are removed from the atmosphere by rainfall, diffusion across the air-sea interface, and chemical degradation. The dominant mechanism for the removal of DDT from the atmosphere is probably rainfall. In England, DDT concentrations in the range from 73 to 210 ppm have been reported in rain in areas close to regions where DDT has been used, and similar concentrations have been reported in the United States (*45*). A DDT concentration in meltwaters from Antarctic ice of 40 ppt has been reported recently (*10*). Earlier measurements were less sensitive (*46*).

DDT concentrations in rainfall vary appreciably throughout the year. The variation is related to the seasonal application of DDT. The fact that there is a seasonal variation suggests that the time constant for removal of DDT from the atmosphere probably does not exceed a few years. If the average DDT concentration in rainfall were 60 ppt and precipitation averaged 1 meter per year, rainfall would remove a total of 3×10^{10} grams of DDT residues from the atmosphere annually, most of that into the oceans. The annual amount of DDT produced throughout the world in the mid-1960's was about 10^{11} grams, approximately 3½ times the amount that would be removed worldwide by rain containing 60 ppt of DDT. Thus an average rainfall concentration of DDT of 60 ppt gives an upper limit for the mean time for removal of DDT residues from the atmosphere by rainfall of about 3.3 years. Residues deposited on the ground are, of course, available for reevaporation.

Measurements of the transfer of carbon dioxide into the oceans suggest a time constant for the downward transport of DDT of 7 years (*47, 48*). This period is extremely long as compared to estimates for diffusive transport. Although one might expect atmospheric DDT to be transferred more slowly than carbon dioxide because of DDT's low solubility in water, direct measurements seem to be lacking. Accumulation of lipids at the ocean surface (*49*) would probably increase the rate of transfer as would association of DDT residues with particles in air. We have assumed a time constant of 4 years for the transfer of DDT residues from the atmosphere to the earth's surface. The mean residence time is probably not longer than this.

Table 1. DDT residues in soils of agricultural and nonagricultural land in the United States. Data were selected because of large sample size or because they are the only data available. For more detailed tabulations see Edwards (*21, 38*).

Soil sites	Sites sampled (No.)	DDT residues (g/m²)		Reference
		Range	Mean	
		Agricultural		
Orchards	14	0.34 –22.1	6.0	(*29*)
Crops	24	0 – 0.87	0.24	(*29*)
Root crops	48	0.045 – 5.73	1.25	(*30*)
Vineyards	2	2.13 – 3.18	2.69	(*31*)
Orchards	2	8.18 –14.60	11.4	(*32*)
Vegetable crops	10	0.07 – 9.52	2.62	(*33*)
Randomly selected	41	0.002 – 1.30	0.148	(*33*)
Alfalfa crops	12	0.06 – 0.98	0.336	(*34*)
Soybean crops	43	0.004 – 4.03	0.986	(*35*)
		Nonagricultural		
Boreal forests (sprayed)	3	0.179 – 0.258	0.213	(*22*)
Boreal forests (unsprayed)			0.0045	(*23*)
Forest in Pennsylvania (unsprayed)		0.0003– 0.0006	0.0004	(*28*)

Chemical degradation of DDT in the atmosphere may also be important. Efficient photodegradation of DDT vapor occurs primarily at wavelengths shorter than 2700 angstroms (*50*). These wavelengths are heavily absorbed by the atmospheric ozone layer. Residues adsorbed on particles are probably considerably more resistant, however. We have assumed atmospheric degradation to be unimportant as compared to transport, but this topic is obviously in need of further study.

DDT in the Oceans

DDT residues circulate initially in the "mixed" layer, which frequently extends to a depth of 75 to 100 meters. They are transferred slowly below the thermocline into the much larger volume of the abyss (*51*). Sedimentation of organic matter removes DDT residues from the upper layers, but direct measurements of sedimentation of DDT residues seem to be lacking. We assume that the virtual insolubility of DDT in water combined with its solubility in fat assures the association of DDT with organic matter and that the rate of transfer of carbon to the abyss would approximate the rate of transfer of DDT residues. Biological mixing may also be important in DDT transport within the ocean, but direct evidence is lacking. As an estimate of the time for the transport of DDT from the mixed layer to the abyss, we have used a result from studies of carbon dioxide that indicate a mean transfer time of about 4 years (*47, 48*).

Within the abyss, transfer rates for carbon dioxide and presumably for other substances such as DDT are very slow indeed, ranging up to hundreds and even thousands of years for certain segments. Because of the fact that the volume of the abyss is immense and because of the possibility that DDT residues may be lost to sedimentation, the abyss is a very large reservoir, virtually infinite for the purposes of this discussion.

DDT in the Biota

The total amount of DDT retained within the biota is small by comparison with the totals that can be retained in other pools within the biosphere. It is also small by comparison with the annual amount of DDT produced. Liberal assumptions with respect to the concentrations of residues in various segments of the biota, including man, lead to an estimate of 5.4×10^9 grams of DDT held within the biota worldwide (Table 2). Estimates of the world biomass are notoriously variable but they are probably correct to within a factor of 2 to 3, almost certainly to within less than a factor of 10. DDT analyses that are specifically appropriate for compilation of such an inventory are few, and the data of Table 2 are, at best, crude estimates. The data on residues have been expressed to orders of magnitude only to avoid a false indication of precision. Questionable estimates have been resolved in the direction of the higher order of magnitude, thus giving a bias toward a higher estimate. The analysis indicates that there may now be between 10^9 and 10^{10} grams of DDT circulating in the biota, about 1/30 of the amount produced in 1 year during the mid-1960's. We consider this an estimate of the maximum amount of DDT that could be in the biota; the only other estimate available is 6×10^8 grams for the biota of the oceans alone published by the M.I.T. study group (*14*). This means that, despite the importance of the biota and the effects of DDT on it, the capacity of the biota for holding DDT residues is small enough that we can ignore it for the moment in our attempt to appraise the worldwide movements of DDT.

Table 2. DDT residues in the biota in the late 1960's. Concentrations are expressed to the nearest order of magnitude only; ppm, parts per million.

Location	Dry biomass ($\times 10^9$ metric tons)	DDT content (ppm)*	Total DDT ($\times 10^8$ g)
	Plant biomass†		
On land			
Lakes and streams	0.04	0.010	0.004
Swamps and marshes	24	0.001	0.240
Terrestrial vegetation (forests, desert, savanna, grassland, tundra)	1814	0.0001	1.814
Agriculture	14	0.1	14.000
Total land	1852		15.058
In oceans			
Open ocean algae	1.0	0.1	1.0
Continental shelf algae	0.3	1.0	3.0
Attached algae	2.0	1.0	20.0
Total ocean	3.3		24.0
Total plants	1855		39.06
	Animal biomass‡		
On land			
Feral mammals	0.009	1.0	0.09
Domestic mammals	0.17	1.0	1.7
Man	0.30§	1.0	3.0
Birds	0.00024	1.0	0.002
In oceans			
Fish	0.65	1.0	6.5
Mammals	0.055	1.0	0.55
Others (protozoa, coelenterates, annelids, nematodes, mollusks, echinoderms, arthropods)‖	3.02	0.1	3.02
Total animals	4.20		14.86
Total DDT in the biota:		5.4×10^9 g	

* Estimates based on values in the literature and the experience of G.M.W. Sources include (*2–7, 11–14, 20–24, 36, 55, 56*) and others. All estimates are to the nearest order of magnitude only; questionable data have been resolved toward the higher number. † Adapted from Whittaker (*60*). ‡ Adapted from Bowen (*61*). § Bowen used 0.03×10^9 tons, which seems to be about 10 times too low (G.M.W.). ‖ Listed by Bowen (*61*); obviously incomplete, but indicative.

A Model of DDT Circulation in the Biosphere

The number of pathways that are clearly important in the worldwide movement of residues appears to be small. The primary reservoirs are the land surface, the troposphere, the mixed layer of the ocean, and the abyss. In the previous sections, we have estimated time constants for the dominant physical processes. The constants have been used in a set of first-order rate equations to yield estimates of DDT loads in the various reservoirs as functions of time. The rate equations have the form:

$$\frac{dN_i}{dt} = R_i(t) - \sum_{j=1}^{m} \frac{N_i}{\tau_{ij}{}^{(l)}} + \sum_{j=1}^{m} \frac{N_i}{\tau_{ij}{}^{(g)}},\ i = 1 \ldots m$$

Here $R_i(t)$ is the rate at which newly produced DDT is introduced into the ith reservoir and N_i is the amount of DDT in the ith reservoir. The sums represent losses from and gains to the ith reservoir; $\tau_{ij}^{(l)}$ is the time constant for DDT loss from the ith to the jth reservoir, and $\tau_{ij}^{(g)}$ represents inputs from the jth to the ith reservoir. There are m reservoirs, and there are thus m simultaneous, first-order, differential equations to be solved.

We have used the time constants estimated above to solve the rate equations over the period from 1940 to 2000 on a digital computer. The DDT input for each year has been taken to be twice the amount produced in the United States per year and has been projected in two ways (Fig. 1, curves A and B). Except for the distinction between land and ocean, no attempt has been made to include geographical variation. Local fluctuations may be expected to be large.

The calculated average DDT concentrations in the atmosphere and in the mixed layer of the oceans for the period from 1940 to 2000 are shown in Fig. 2. If the world DDT production becomes zero in 1974, the concentration in the lower atmosphere would have reached a peak in 1966 at about 72 ppt (84×10^{-9} gram per cubic meter). The mixed layer of the ocean would contain its maximum of 15 ppt in 1971. The concentrations in both reservoirs can be expected to decline gradually, with the concentration in air reaching 10 percent of its peak value in 1984. The concentration in the mixed layer will not decline to 10 percent of its peak value until 1993. The total load of DDT on the land surface reached a maximum concentration of 6.34×10^{11} grams in 1964 to 1966, and will decline if DDT production slows.

There is reason to assume that the worldwide production of DDT will not drop, but may increase, despite U.S. restrictions. The increase will be in response to an increasing demand for an inexpensive means of pest control in agriculture and for control of vectors of disease, especially malaria. If we assume that foreign production replaces U.S. production and that the total use of DDT in the world increases after 1969 (curve B, Fig. 1), the concentrations of DDT in air and water will follow the curves marked B in Fig. 2, continuing to increase until after the year 2000.

Implications for Life

The physical processes we have discussed dominate the transfer of DDT residues throughout the biosphere. Living systems retain quantities of DDT that are small by proportion, and living things appear, at least superficially, to play a minor role in the world budget of DDT. Yet it is the residues that are available to living systems that are the hazard, and we must examine their behavior with special care. The total quantities of DDT residues in the biota are but 1/30 or less of the annual amount of DDT produced in recent years; they are also a small fraction of the annual transfers estimated from soils to air and the oceans. The quantities are small enough that the transfer from land to water by surface runoff, small as it is, must be assumed to contribute to contamination of the coastal biota. The residues presently held in the biota, and the maximum quantity that the biota could hold (not very greatly different), are so small in proportion to the total amount of DDT produced that we wonder why the biota has not been affected much more drastically than it has been—and what the future holds.

The answers are far from clear. DDT residues are accumulated in living systems and recycled in much the same ways that certain elements essential for life are recycled. Just as phosphorus is recycled from sediments by various means, so DDT residues may reenter complex food webs from organic sediments. One such route is by direct consumption of detritus (*52*). Others must include oxidation of the sediments. Concentrations of DDT in the biota may ultimately reach as much as 10^6 times the concentrations in the general environment (*36*). The effects of high concentrations are clear enough: food webs are reduced, carnivores eliminated, and hardy, small-bodied organisms favored (*53*). The changes are similar to those that occur in eutrophication; the sedimentation of organic matter is probably increased, often speeded by a shift toward an increasingly anaerobic benthos. We assume that under such extreme conditions DDT residues tend to accumulate in anaerobic sediments and are removed from circulation. Thus one of the effects of DDT is to reduce the biota and to increase the rate of removal of DDT residues into sediments. The process tends to restrict the movement of residues in the larger circulations of the biosphere, accentuating the importance of local contamination. There is, however, not much question that the oceans, as well as lakes and estuaries, are vulnerable to such effects. How much more DDT would it take to degrade the biota significantly?

The answer hinges on both the rate of movement of residues through the major reservoirs of the biosphere and on the coupling between the biota and the environment. How rapidly does the biota absorb DDT? Is there a possibility of the biota's achieving an equilibrium in which inputs of DDT residues are exactly balanced by losses?

DDT residues enter the biota both through food webs and by direct absorption. The relative importance of these routes varies between land and water and among species, and the time for the biota to come to equilibrium with residues in the environment must also vary. One attempt at appraising the time for a food web to reach equilibrium led to an estimate of between "four times the average life span of the longest-lived species and the sum of the life spans for all trophic levels" (*54*, p. 506). Such an analysis suggests that equilibrium for the entire biota would be reached only after many decades. Movement of DDT residues into the abyss appears much more rapid than this.

On the other hand, plankton in water would be expected to reach equilibrium with residues in solution in the water very rapidly, and small-bodied, warm-blooded carnivores that have high rates of metabolism and feed from water-based food webs might be expected to accumulate high concentrations of residues rapidly and to be affected by them. This circumstance, of course, is what we see: aquatic carnivorous birds accumulate high concentrations of DDT and then their numbers rapidly diminish. So, although the biota as a whole may not have achieved equilibrium with the residues circulating now, certain segments of the biota are being reduced, a fact that indicates that the biota may be appreciably changed before an "equilibrium" with present rates of DDT production and present world inventories is reached. Under these circumstances the concept of an equilibrium becomes elusive; there is no true equilibrium, only a constant state of flux through a pool that probably grows smaller as the biota is reduced. The coupling between DDT residues in the environment and in aquatic food webs would seem to be reasonably close, especially for lower trophic levels, and a decline in the amount of DDT used should be reflected almost immediately in lower concentrations in the biota.

A variety of evidence favors this conclusion. Fauna of salmon streams in the Miramichi River in New Brunswick, Canada, and fish in Sebago Lake, Maine, responded year by year to a reduction in the use of DDT (*55*). More recent experience on Long Island with osprey populations seems to be indicating a similar response, although the observations are far from conclusive. Populations dropped abruptly during the mid-1960's. The decline seems to have been arrested, perhaps reversed, after cessation of use of DDT for mosquito control in 1967 (*56*). Terrestrial ecosystems probably respond more slowly, but the patterns of circulation of residues and their effects are similar.

These examples and others, such as the observation that the effects of pesticides on reproduction of bird populations in England are related to the intensity of use of chlorinated pesticides (*57*), emphasize that most conspicuous effects on the biota in the past have not been the result of worldwide movements of DDT residues but rather have been attributable to local concentrations identifiable with some local or regional use. Restriction in the use of pesticides has usually reduced the effects within a few years.

Residues in other segments of the biota, however, are very much more closely related to larger patterns of circulation. Oceanic birds such as the sooty and slender-billed shearwaters (*4, 5*) and the Bermuda petrel (*6*) that feed in the open ocean, the California mackerel, the penguin, and the crabeater seal of the Antarctic (*58*) must obtain their residues from patterns of circulation that are close to being "worldwide." All of these organisms are contaminated with DDT, some with concentrations occasionally exceeding 10 ppm. These are the organisms that are most closely coupled to the major nonliving pools of DDT circulating in the biosphere. Our analyses suggest that such organisms should reflect world use of DDT within a few years, with residues in the biota increasing or decreasing as world use rises or falls. The fact that none of these organisms has yet become extinct from DDT effects is mere good fortune: the total amounts of DDT estimated to be circulating in the biosphere are many times greater than the amounts required to eliminate most such animals. We know that the residues can be concentrated by many factors of 10 into the biota—and affect it catastrophically. Yet, although there is no question about the devastation wrought by DDT locally and even regionally, the worldwide component seems not yet to have reached the point of widespread extinctions. (The difficulties of measuring effects are so great as to make most biologists who examine this question in any depth suspicious that effects may be occurring unobserved or masked by other causes). If we assume that the lower trophic levels of aquatic food webs are more closely coupled to their environment by dint of the two pathways for entry of residues than the analyses of Harrison *et al.* (*54*) suggest, then reduction in the use of DDT should be reflected within a few years in a reduction in the DDT residues in the biota identified with the worldwide distribution.

Where Has the DDT Gone?

The physical and chemical characteristics of DDT might lead one to assume that the biosphere should behave as a giant separatory funnel, gradually partitioning the lipid-soluble residues into the lipid-rich biota. Although there is no question that this process does occur, there is also no escape from the conclusion that it does not work well on the biospheric level. Most of the DDT produced has either been degraded to innocuousness or sequestered in places where it is not freely available to the biota. Recent work seems to support the latter assumption and the assumptions of our model. A preliminary report of detailed analyses of DDT residues in the air of nine U.S. cities in 1967–1968 (*59*) shows concentrations in winter, when DDT is not used locally and residues might be expected to be mixed throughout the troposphere, commonly falling between 10^{-9} and 100×10^{-9} gram per cubic meter. The range approximates our prediction of 84×10^{-9} gram per cubic meter in air based on the assumption of declining use. The observation supports our assumptions on the routes of movement and sizes of pools. The fact remains, however, that, despite the abundance, persistence, and worldwide distribution of DDT residues, they are not as freely available to the biota as might be assumed. How and precisely where they are held is not yet clear, but the biosphere appears to have a large capacity for holding them apart from the biota. What is clear is that large quantities of DDT were introduced into use before any appraisal was made of the capacity of the biosphere for receiving them. In this instance man seems to have been blessed with extraordinary good fortune.

Summary

The worldwide pattern of movement of DDT residues appears to be from the land through the atmosphere into the oceans and into the oceanic abyss. Calculations based on the fragmentary data available on rates of movement and sizes of various pools of DDT residues lead to the conclusion that concentrations in the atmosphere and in the mixed layer of the oceans lag by only a few years behind the amounts of DDT used annually throughout the world. A model suggests that maximum concentrations of DDT residues occurred in air in 1966 and will occur in the mixed layer of the oceans in 1971. The biota probably contains in total less than 1/30 of 1 year's production of DDT during the mid-1960's, a very small amount in proportion to the total potentially available. The reason for the biota's failure to absorb larger quantities and to be affected much more severely is unclear. The analysis suggests that mere good fortune has protected man and the rest of the biota from much higher concentrations, thus emphasizing the need to determine the details of the movement of DDT residues and other toxins through the biosphere and to move swiftly to bring world use of such toxins under rational control based on firm knowledge of local and worldwide cycles and hazards.

References and Notes

1. DDT residues include DDT and its decay products, DDD and DDE: DDT, 1,1,1,-trichloro-2,2-bis(*p*-chlorophenyl)ethane; DDE, 1,1 dichloro-2,2-bis(*p*-chlorophenyl)ethylene; DDD, 1,1-dichloro-2,2-bis(*p*-chlorophenyl)ethane; dieldrin, 1,2,3,4,10,10-hexachloro-6,7-epoxy-1,4,-4a,5,6,7,8,8a-octahydro-endo-exo-1,4:5,8-dimethanonaphthalene.
2. For a discussion of residues in man, see U.S. Department of Health, Education and Welfare, *Report of the Secretary's Commission on Pesticides and Their Relation to Environmental Health* (Government Printing Office, Washington, D.C., 1969).
3. Some of the most recent studies establishing the extent of contamination are: R. W. Risebrough (*4*); ———, D. B. Menzel, D. J. Martin, H. S. Olcott (*5*); C. F. Wurster, Jr.,

and D. B. Wingate (*6*); E. C. Tabor (*7*).

4. R. W. Risebrough, in *Chemical Fallout*, M. W. Miller and G. G. Berg, Eds. (Thomas, Springfield, Ill., 1969), p. 5.
5. ———, D. B. Menzel, D. J. Martin, H. S. Olcott, *Nature* **216**, 589 (1967).
6. C. F. Wurster, Jr., and D. B. Wingate, *Science* **159**, 979 (1968).
7. E. C. Tabor, *Trans. N.Y. Acad. Sci.* **28**, 569 (1966).
8. P. Antommaria, M. Corn, L. DeMaio, *Science* **150**, 1476 (1965); D. C. Abbott, R. B. Harrison, J. O. Tatton, J. Thompson, *Nature* **211**, 259 (1966).
9. R. W. Risebrough, R. J. Huggett, J. J. Griffin, E. D. Goldberg, *Science* **159**, 1233 (1968).
10. T. J. Peterle, *Nature* **224**, 620 (1969).
11. N. C. W. Moore, Ed., *J. Appl. Ecol.* **3** (Suppl.), xxx (1966).
12. G. M. Woodwell, *Sci. Amer.* **216**, 24 (March 1967).
13. J. Frost, *Environment* **11**, 14 (1969).
14. Massachusetts Institute of Technology, *Man's Impact on the Global Environment: Report of the Study of Critical Environmental Problems* (M.I.T. Press, Cambridge, Mass., 1970), pp. 126–131.
15. In the United States the Secretary of Health, Education, and Welfare, the Secretary of Interior, the Secretary of Agriculture, and the administrator of the Environmental Protection Agency have moved recently toward removing DDT from most uses by 1972. These moves are being challenged, and the question remains unresolved. Certain other countries, including Canada and England, have restricted use of persistent pesticides. There is no basis for expecting an international ban and little basis for expecting restraint in the use of DDT outside of these countries.
16. G. M. Woodwell, *BioScience* **19**, 884 (1969).
17. B. Commoner, *Science and Survival* (Viking, New York, 1963).
18. M. C. Bowman, F. Acree, Jr., M. K. Corbett, *Agr. Food Chem.* **8**, 406 (1960).
19. T. F. West and G. A. Campbell, *DDT and Newer Persistent Insecticides* (Chemical Publishing Company, New York, 1952).
20. There is no completely satisfactory way of appraising the persistence of DDT in nature; residues have different degrees of persistence in different places. The concept of "half-life" implies a systematic degradation that may not occur universally. S. G. Herman, R. L. Garrett, and R. L. Rudd [in *Chemical Fallout*, M. W. Miller and G. G. Berg, Eds. (Thomas, Springfield, Ill., 1969), p. 24] show that the population of western grebes of Clear Lake, California, included individuals having substantially the same range of concentrations of DDD residues over a 10-year period despite the cessation of spraying; mean values dropped by about one-half in that period. Residues are known to remain for many years in soil, especially in organic soils. See the review by C. A. Edwards (*21*); see also G. M. Woodwell and F. T. Martin (*22*), J. Dimond (*23*), and R. G. Nash and E. A. Woolson (*24*). These observations all suggest that residues persist with mean lives of many years. The assumption of a 10-year half-life for residues within the biosphere as a whole has long appeared reasonable to one of us (G.M.W.) (*16*); the assumption may be error in that residues tend to be stored or cycled in places where they are not degraded chemically and may persist longer.
21. C. A. Edwards, *Residue Rev.* **13**, 83 (1966).
22. G. M. Woodwell and F. T. Martin, *Science* **145**, 481 (1964).
23. J. Dimond, *Maine Agr. Exp. Sta. Misc. Rep. 125* (1969).
24. R. G. Nash and E. A. Woolson, *Science* **157**, 924 (1967).
25. U.S. Tariff Commission, *Synthetic Organic Chemicals, U.S. Production and Sales, 1969* (Government Printing Office, Washington, D.C., 1970); *U.S. Dep. Agr. Econ. Rep. No. 158* (April 1969).
26. R. Carson recognized in 1963 the significance of aerial transport of DDT residues in contaminating the oceans. She cited observations in Maine and elsewhere indicating that only half the spray emitted from aircraft lands on the ground, the rest being dispersed in the atmosphere. Recent measurements of DDT attached to particulate matter in the atmosphere confirm the earlier conclusions (R. Carson, statement to the Subcommittee on Reorganization and International Organizations of the Committee on Government Operations, U.S. Senate, May–June 1963, part 1, p. 207); G. M. Woodwell, *Forest Sci.* **7**, 194 (1961); R. W. Risebrough, R. J. Huggett, J. J. Griffin, E. D. Goldberg (*9*); O. B. Cope, *Trans. Amer. Fish. Soc.* **90**, 239 (1961).
27. Most measurements have been in agricultural soils where there was a real question about hazards after long use. Published data are, therefore, heavily skewed toward high values, and any tabulation such as that of Table 1 may be misleading. The data of Table 1 are representative of published reports: H. Cole *et al.* (*28*); E. P. Lichtenstein (*29*); W. L. Seal *et al.* (*30*); E. F. Taschenberg *et al.* (*31*); L. C. Terriere *et al.* (*32*); W. L. Trautman *et al.* (*33*); G. W. Ware *et al.* (*34*); U.S. Department of Agriculture (*35*). The maximum in this tabulation was in an orchard in Indiana that contained DDT residues totaling 22.0 grams per square meter. Almost all agricultural soils contain detectable DDT residues, most in excess of 0.056 gram per square meter. Averages of several samples from a region are rarely less than 0.224 gram per square meter, according to this tabulation. Nonetheless, Table 1 contains few data from crops that are rarely sprayed, such as many grains and fodder crops. Any average calculated for agricultural soils must account for the extensive acreages devoted to these crops as well. The average contamination of agricultural land is probably greater than the mean reported for 41 randomly sampled agricultural and forest soils in Wisconsin and eight states west of the Mississippi River (Table 1). In that tabulation, 22 of the soils contained less than 0.0022 gram of DDT per square meter, but others had high enough concentrations to produce an average concentration of 0.148 gram per square meter. It seems very unlikely that the average amount of DDT in agricultural soils, including all agricultural soils, would approach the value of 0.99 gram per square meter reported for soybeans in 1968 (Table 1). A reasonable estimate of the average value for agricultural soils where DDT is used would seem to be in the range of 0.140 to 0.56 gram per square meter. The average would be skewed toward the lower end of the range by the inclusion of crops that are sprayed irregularly. We have assumed for our calculations an average contamination of agricultural soils of 0.168 gram per square meter.
28. H. Cole, D. Barry, D. E. H. Frear, A. Bradford, *Environ. Contam. Toxicol.* **2**, 127 (1967).
29. E. P. Lichtenstein, *J. Econ. Entomol.* **50**, 545 (1957).
30. W. L. Seal, L. H. Dawsey, G. E. Cavin, *Pesticide Monit. J.* **1**, 22 (1967).
31. E. F. Taschenberg, G. L. Mark, F. L. Gambrell, *Agr. Food Chem.* **9**, 207 (1961).
32. L. C. Terriere, U. Kiigemagi, R. W. Zwick, P. H. Westigard, in *Organic Pesticides in the Environment*, R. F. Gould, Ed. (American Chemical Society, Washington, D.C., 1966), p. 263.
33. W. L. Trautman, G. Chesters, H. B. Pionke, *Pesticide Monit. J.* **2**, 93 (1968).
34. G. W. Ware, B. J. Estensen, W. P. Cahill, *ibid.*, p. 129.
35. U.S. Department of Agriculture, Plant Pest Control Division, Agricultural Research Service, *ibid.*, p. 58.
36. Most of the nonagricultural soils for which data on DDT residues are available are also soils from areas that have received repeated applications of DDT. The highest values range from several tenths of a gram per square meter for a marsh along the eastern coast [G. M. Woodwell, C. F. Wurster, Jr., P. A. Isaacson, *Science* **156**, 821 (1967)] and for other organic soils that had been heavily treated, to zero, or at least to values below the limits of detection. Unsprayed forested areas in Pennsylvania (Table 1) contained in 1967 4.5×10^{-4} gram per square meter. Soils in Maine forests that had never been sprayed contained ten times that quantity (Table 1). Organic soils and sediments contain more residues, but usually not more than a few tenths of a pound per acre. Any world average estimated from such data is most tenuous. We estimate that nonagricultural soils contain a minimum of 1.1×10^{-4} gram per square meter. A reasonable estimate of the mean concentration of DDT in the temperate zone of the Northern Hemisphere appears to be 4.5×10^{-4} gram per square meter as reported for the soils of Pennsylvania forests (Table 1).
37. *Hammond Citation World Atlas* (Hammond, Maplewood, N.J., 1966), p. 192.
38. C. A. Edwards, *Persistent Pesticides in the Environment* (Chemical Publishing Company, New York, 1970).
39. G. S. Nazarov, *Tr. Saratov. Zootekh. Vet. Inst.* **7**, 319 (1958).
40. W. F. Spencer and M. M. Cliath, *Environ. Sci. Technol.* **3**, 670 (1969).
41. "Net primary production" usually refers to the net amount of dry matter produced per unit of land are → G. M. Woodwell and R. H. Whittaker, *Amer. Zool.* **8**, 19 (1968); E. P. Odum, *Fundamentals of Ecology* (Saunders, Philadelphia, ed. 3, 1971); G. M. Woodwell, *Sci. Amer.* **223**, 64 (September 1970).
42. L. H. Long, Ed., *World Almanac* (Newspaper Enterprise Association, Inc., New York, 1967), p. 273; A. M. Piper, *U.S. Geol. Surv. Water Supply Pap. No. 1797* (1965).
43. L. Weaver, C. G. Gunnerson, A. W. Breidenbach, J. J. Lichtenberg, *Public Health Rep.* **80**, 481 (1965).
44. A. Standen, Ed., *Encyclopedia of Chemical Technology* (Interscience, New York, ed. 2, 1966), vol. 11, p. 691.
45. K. R. Tarrant and J. O. G. Tatton, *Nature* **219**, 725 (1968); G. A. Wheatley and J. A. Hardman, *ibid.* **207**, 486 (1965).
46. J. L. George and D. F. H. Frear, *J. Appl. Ecol.* **3** (Suppl.), 155 (1966).
47. H. Craig, *Tellus* **9**, 1 (1957).
48. R. Revelle and H. E. Suess, *ibid.*, p. 18.
49. W. D. Garrett, *Deep-Sea Res.* **14**, 221 (1967); D. B. Seba and E. F. Corcoran, *Pesticide Monit. J.* **3**, 190 (1969).
50. H. Lipne and C. W. Kearns, *J. Biol. Chem.* **234**, 2129 (1959); L. Goldberg, in *The Earth as a Planet*, G. P. Kuiper, Ed. (Univ. of Chicago Press, Chicago, 1954), p. 434; L. R. Koller, *Ultraviolet Radiation* (Wiley, New York, ed. 2, 1965); A. R. Mosier, W. D. Guenzi, L. L. Miller, *Science* **164**, 1083 (1969); N. Bhandari, D. Lal, Rama, *Tellus* **18**, 391 (1966).
51. W. Wooster, quoted by H. Craig (*47*).
52. W. E. Odum, G. M. Woodwell, C. F. Wurster, *Science* **164**, 576 (1969).
53. G. M. Woodwell, *ibid.* **168**, 429 (1970).
54. H. L. Harrison, O. L. Loucks, J. W. Mitchell, D. F. Parkhurst, C. R. Tracy, D. G. Watts, V. J. Yannacone, Jr., *ibid.* **170**, 503 (1970).
55. M. H. A. Keenleyside, *Can. Fish Cult.* **24**, 17 (1959); *J. Fish. Res. Board Can.* **24**, 807 (1967); F. P. Ide, *ibid.*, p. 769; R. B. Anderson and W. H. Euerhart, *Trans. Amer. Fish. Soc.* **95**, 160 (1966); R. B. Anderson and O. Fenderson, *J. Fish. Res. Board Can.* **27**, 1 (1970); R. B. Anderson, *Sebago's Bright Future* (Maine Department of Inland Fisheries and Game, Augusta, 1966); S. DeRoche, unpublished data on fish size in Sebago Lake, 1957–1967, taken by the Maine Department of Inland Fisheries and Game; R. L. Rudd, *Pesticides and the Living Landscape* (Univ. of Wisconsin Press, Madison, 1964).
56. Counts of nesting pairs and young by D. Puleston [Brookhaven Lecture Series No. 104, *Brookhaven Nat. Lab. Publ. 50309* (15 September 1971)] and his colleagues over more than 20 years have documented the catastrophic decline of the osprey in the early 1960's, and the slight recovery of reproductive success in recent years. Whether the small residual population will recover remains doubtful, however.
57. D. A. Ratcliffe, *J. Appl. Ecol.* **7**, 67 (1970).
58. W. J. L. Sladen, C. M. Menzie, W. L. Reichel, *Nature* **210**, 670 (1966).
59. C. W. Stanley, J. E. Barney, II, M. R. Helton, A. R. Yobs, *Environ. Sci. Technol.* **5**, 430 (1971).
60. R. H. Whittaker, *Communities and Ecosystems* (Collier-Macmillan, New York, 1970).
61. H. J. M. Bowen, *Trace Elements in Biochemistry* (Academic Press, New York, 1966).
62. Research was carried out under the auspices of the U.S. Atomic Energy Commission.