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34] Pathways of Pesticide Dispersion in the Environment

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ABSTRACT

Pesticide dispersion is a natural consequence of the Second Law of Thermodynamics; once applied to an agricultural field, pesticides will migrate in a manner tending to create equal fugacity in all phases of the global environment. On a local scale, this movement is somewhat predictable, given a knowledge of chemical properties, weather, soil type, formulation, and details of the application. Pesticide movement on a regional or global scale is more difficult to predict, but is known to occur. The pathways by which chemicals disperse are controlled by both intrinsic chemical properties and extrinsic environmental factors. Because of the heterogeneous and changeable character of natural water and the atmosphere, the extrinsic factors are difficult to research and model. This report examines the interaction of chemical and environmental factors that control local, regional, and global pesticide dispersion.

INTRODUCTION

In 1965, the U.S. Department of the Interior made the dramatic announcement that DDT residues had been discovered in Antarctica (Sladen, Menzie, and Reichel 1966). This was astonishing news, since it seemed inconceivable at the time that DDT should be present in a pristine wilderness so far from any possible use of the chemical. The vanishingly small vapor pressure and solubility implied that DDT was immobile, and no mechanism appeared to exist by which it could move away from the site of application.

Such early evidence of the widespread environmental contamination by DDT and other chlorinated pesticides was greeted with intense skepticism. In the

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Table 34.1. Intrinsic and Extrinsic Factors That Control Pesticide Dispersion in the Environment

Intrinsic Properties
Vapor pressure
Solubility
Resistance to chemical change
Bioactivity
Derived Properties
Hydrophobicity (K_{ow})
Soil/Water partitioning (K_{oc})
Bioconcentration
Air/Water partitioning (H)
Extrinsic Factors
Formulation/Management
Weather (rain, solar energy)
Inhomogeneity (temperature, moisture, organic matter)
Heterogeneity of air and water
Kinetics (mixing times, residence times, diffusion-controlled processes).

intervening years, through the efforts of many scientists, we have documented not only local and regional pesticide movement, but have also expanded our list of chemicals—both pesticides and industrial chemicals—that are dispersed on a global scale. We now know that the presence of DDT in Antarctica resulted as a natural consequence from the properties of DDT. Although it has a low vapor pressure and persists in soil for years, it will volatilize—from cotton plants, for example, with a 50% disappearance time of just over 10 days (Willis et al. 1983)—and thereafter be carried great distances in the atmosphere. It is finally brought to the surface by precipitation. DDT entered Antarctica on snow.

We now more clearly understand that a pesticide-treated field represents a highly ordered state that tends toward disorder—consonant with the Second Law of Thermodynamics—by dispersion of pesticide into the surrounding environment through the fluid components water and air. Great concentration differences exist between the treated site and the corresponding components of the surrounding environment. For example, close to a treated soil surface an evaporating pesticide may establish a vapor density approaching its own characteristic saturation vapor concentration. Air flowing over the field contains essentially no pesticide. As the pesticide-laden air near the surface becomes mixed in the flowing stream, dilution occurs, and there is very little tendency for the pesticide to return to the surface.

Once the air moves beyond the boundary of the treated field, the situation is reversed. There is now no pesticide at the surface, and there then exists a finite tendency for the pesticide to migrate from the flowing air to the surface. This is a manifestation of the fact that pesticide movement will occur in directions that will reduce concentration differences, eventually leading to equilibrium among all phases of the global environment.

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Table 34.2. Intrinsic and Derived Properties of Heptachlor and Atrazine

Property	Heptachlor	Atrazine
Vapor pressure (mm Hg)	3×10^{-4}	5.6×10^{-7}
Solubility (ppm)	0.056	33
Soil half-life	1 year	35 days
Soil/Water partitioning Coefficient (K_{oc})	2×10^4	149
Bioconcentration factor-static-	17,400	0
flowing-	2,150	11
Air/Water partitioning Coefficient (H)	0.11	2×10^{-7}

This movement and its environmental consequences will reflect, in the first analysis, the intrinsic properties of the chemicals (Table 34.1): water solubility, vapor pressure, resistance to chemical alteration, and biological activity. Through these, transport tendencies are identified through derived properties: hydrophobicity (tendency to partition between water and organic matter), soil/water partitioning (tendency to leach, move in runoff, be adsorbed to sediments), bioconcentration factors (tendency to partition between water and fat or protein), and Henry's Law behavior (tendency to partition between water and air). These derived properties tell us the direction of migration by identifying the most likely distribution when the pesticide reaches global equilibrium.

They are not the entire story, however. The amount of pesticide transported to the field boundary, and especially the rate at which the movement occurs, is greatly modified by such factors as formulation, management, weather, and moisture and temperature status at the field or crop surface. Some of these extrinsic factors are listed in Table 34.1. To illustrate how intrinsic and extrinsic factors influence dispersion, I will compare environmental data on two very different pesticides, heptachlor and atrazine.

Heptachlor is a chlorinated hydrocarbon once used as a soil insecticide in corn (now banned). Atrazine is a s-triazine herbicide widely used to control broadleaf weeds in corn. Both are moderately persistent in soil. They are markedly different in the key intrinsic properties of vapor pressure and solubility; heptachlor is hydrophobic and volatile, while atrazine has less tendency to associate with organic matter or escape to the atmosphere but a great tendency to move in water (Table 34.2). Together, their properties span those of many pesticides.

MOVEMENT IN WATER

The water environment is very heterogeneous. All natural water contains, in addition to suspended solids, dissolved or colloidal organic matter, which is known to interact strongly with pesticides in solution. Some of this organic matter, primarily lipid and decaying proteinaceous material, accumulates at the air/water interface, creating surface microlayers in which hydrophobic pesticides may accumulate. In addition to these organic phases, pesticides partition to

suspended solids, sediment, and biota. They also reversibly partition between water and air through surface accumulation, evaporation, deposition, and bubble ejection.

An excellent review of pesticide movement in runoff was given by Wauchope (1978). In general, pesticide concentration in runoff, and the amount lost, depend strongly upon the delay in time between application and first runoff. A linear plot of logarithm of pesticide concentration in runoff against time after application gives excellent agreement to field data for many pesticides. For example, atrazine has a runoff "half-life" of seven to ten days. A "critical" event, as defined by Wauchope, is one with more than one centimeter of rain, half or more of which emerges as runoff, within two weeks of application. Such events always produce the bulk of runoff losses for a season. Thus, weather is an extremely important external factor.

We recently completed a 3-year study of the movement of atrazine and simazine from a 2750 hectare watershed into Wye River, a tributary of Chesapeake Bay (Glotfelty et al. 1983). Not too surprisingly, we found that herbicide transport into the estuary was predominantly by water and that atrazine concentrations in runoff from these coastal plains soils peaked at about 300 ppb when it occurred within two weeks of application. In 1981, most of the corn in the watershed was planted in a brief period of good weather that was followed by "critical" runoff event. We found that 2%–3% of the atrazine applied in the watershed moved to the estuary. In other years when runoff was delayed longer or was less abundant, much smaller amounts reached the estuary.

Another interesting external factor must be added, however. In several years we observed a late-summer enhancement of simazine runoff. In some cases this was very pronounced and occurred even after simazine levels in the soil had declined to low values. We think this can be explained by a wick mechanism, and perhaps an association with organic material.

During a dry period, water evaporation causes bulk moisture in the soil to flow toward the surface. Simazine, along with other soluble organic and inorganic compounds in the soil solution, is thus brought to the surface. This is a mechanism by which some incorporated pesticides evaporate from the soil (Hartley 1969), but simazine accumulates at the surface due to its low volatility. It is then ready to move, perhaps associated with other organics, when runoff occurs.

In contrast to the abundance of data on atrazine runoff, we were unable to locate any data on heptachlor runoff in the open literature. We attempted some years ago to measure heptachlor runoff from a corn field in Ohio, but the data are unpublished because we had sampling problems. They do show what one might expect, and can be compared to similar studies of dieldrin runoff (Caro and Taylor 1971). Losses are a very small fraction of the total treatment, and movement is primarily with eroded soil. Once beyond the field boundary, dieldrin (and we presume heptachlor) quickly moved to the stream sediment, and concentrations were very low a short distance downstream.

Atrazine moved to the estuary with fresh water. When this mixed with the saline water in the estuary, atrazine diluted in a conservative manner. Thus, atrazine concentrations declined linearly with increasing salinity and approached zero concentration at the salinity of the largely uncontaminated open bay. This was because atrazine moves primarily (70%–90%) in the aqueous phase and shows little tendency to adsorb to suspended solids or bottom sediments. However, even atrazine is reported to be strongly associated with colloidal organic material in the estuary (Means and Wijayarante 1982), and we believe this organic material may profoundly affect its aqueous chemistry and accelerate its rate of degradation.

Table 34.3. Partition Coefficients for Accumulation at the Air/Water Interface and Their Relation to Henry's Law Distribution for Selected Pesticides

Insecticide	$K_p \left(\frac{\text{moles/cm}^2}{\text{moles/cm}^3} \right) \times 10^3$	$\frac{H}{K_p}$
Heptachlor	4	30
DDT	8	0.25
Lindane	1.2	0.06

In an open estuary, atrazine shows only a small tendency to accumulate in the surface microlayer (Wu et al 1980). In about half the microlayer samples, atrazine had no significant enrichment, and no enrichment factor was greater than 110, based upon the difference in concentration between the bulk water and a 150 μm layer at the surface. By contrast, enrichment factors exceeding 10^5 have been reported for organochlorine insecticides in marine surface microlayers (Seba and Corcoran 1969).

Because of their hydrophobicity, heptachlor and other organochlorine insecticides tend to accumulate at the air/water interface, even when surface films are absent. In fact, there is a definite three-phase equilibrium established between the liquid, surface interface, and air. Table 34.3 gives values for the partition coefficient K_p between surface and bulk liquid ($K_p = \text{moles cm}^{-2}/\text{moles cm}^{-3}$), and the relation between K_p and Henry's Law coefficients for heptachlor, DDT, and lindane.

One interesting result of this behavior is that the total distribution coefficient of these insecticides on air bubbles in water depends on the bubble surface to volume ratio and that their rate of extraction from water by bubbles and injection thereby into air greatly exceeds that predicted from their Henry's Law coefficients alone. For example, about 67% of the heptachlor associated with a 1-mm air bubble is on the bubble surface; for DDT, the ratio is greater than 99%. The same result is true for drops of water in air.

MOVEMENT IN AIR

Atmospheric transport and return to the surface are largely controlled by the distribution of the pesticide between the vapor phase and airborne particles. Pesticides enter the atmosphere as vapor, on soil dust, as particles of formulation, or by drifting spray droplets. Once in the atmosphere, vapors may be adsorbed or released from particles, or otherwise interact with the atmospheric aerosol, which is extremely heterogeneous. Both particles and vapor may deposit directly at the surface or be brought down in rain, but each does so at distinctly different rates and probably moves to different components of the environment. We are only beginning to see progress in this very complicated area of research. Pesticide transport in air is important to understand, for it may reach global proportions.

The techniques one uses to measure, in the field, post-application pesticide losses to the atmosphere were described in pioneering work of Parmele, Lemon,

and Taylor (1972). We first used these techniques to measure the volatilization losses of heptachlor and dieldrin from cornfields in Ohio (Taylor et al. 1976).

Heptachlor moves to the atmosphere entirely in the vapor phase. Volatilization losses following incorporation of 5.6 kg/ha to the 7.5 cm depth in soil peaked in mid-summer at 5 g/ha/day. Between May and October, losses totaled 6.8% of the amount applied. (Dieldrin losses measured in the same experiment were 3.6% of application. This greatly exceeded typical seasonal losses due to runoff; see Caro and Taylor 1971). The highest observed heptachlor concentration was 0.6 $\mu\text{g}/\text{m}^3$, which was measured close to the soil surface in calm air within the mature corn canopy.

When heptachlor is applied to the surface of moist soil, or to vegetation, volatilization losses are much greater—half within 6 hours and 90% within six to seven days (Glottfelty 1981; Taylor et al. 1977). We have observed periods, both over vegetation and moist soil, in which losses were unrestrained and measured air concentrations of components of an applied mixture were proportional to their respective saturation vapor densities. In fact, in one instance over vegetation, air concentrations could not have been higher had the entire field surface been uniformly coated with the pure chemical. Such high volatilization rates usually occur right after application. In the longer term, losses from moist soil become controlled by diffusion from the thin layer of soil penetrated by the spray.

Solar energy input and air turbulence are typically greatest at noon. It is common to see pesticide volatilization follow this diurnal pattern, peaking at noon and becoming very low at night. But if the soil forms a dry surface layer, volatilization losses are greatly reduced, and they may be dominated by nighttime losses when dew or capillary flow remoistens the soil surface. Long-term losses depend most upon moisture supply to the soil surface and hardly at all on energy input. Specific volatilization rates of heptachlor, trifluralin, lindane, and chlordane ranged from five to forty times less over very sandy soil that formed a dry surface layer, compared with a silt-loam soil that maintained moisture at the surface.

Figure 34.1 shows an interesting variation of this effect. It compares the vertical fluxes of atrazine and alachlor on the third day following their application to bare soil. Atrazine was formulated as a wettable powder and alachlor as an emulsifiable concentrate. They were applied in the same spray. Their fluxes, which were measured essentially over dry soil, show completely opposite behavior.

In the early morning, when the soil was still moist with dew from the previous evening, alachlor flux was high. At noon, when the surface was dry, alachlor flux was low. Since alachlor moves predominantly as vapor, this is the dry soil effect noted above—losses are dependent upon supply of moisture to the surface.

Conversely, from low morning values, atrazine fluxes increased with increasing wind speed and peaked close to noon. This pattern, plus the fact that atrazine in air was trapped quantitatively on glass fiber filters, strongly suggests that atrazine was moving predominantly by wind erosion of particles, probably the wettable powder formulation.

This interpretation is supported by other measurements. Simazine was also applied as a wettable powder in the same spray. Although it is much less volatile than atrazine, its measured vertical fluxes to the atmosphere over this day were virtually the same. Thus, losses were proportional to application rates and not vapor pressures. All simazine in sampled air was trapped on glass-fiber filters, and noontime air concentrations exceeded the known saturation vapor density of the chemical. Thus, atrazine and simazine were lost by wind erosion of the wettable powder formulation in which they were applied. These losses were on the order of

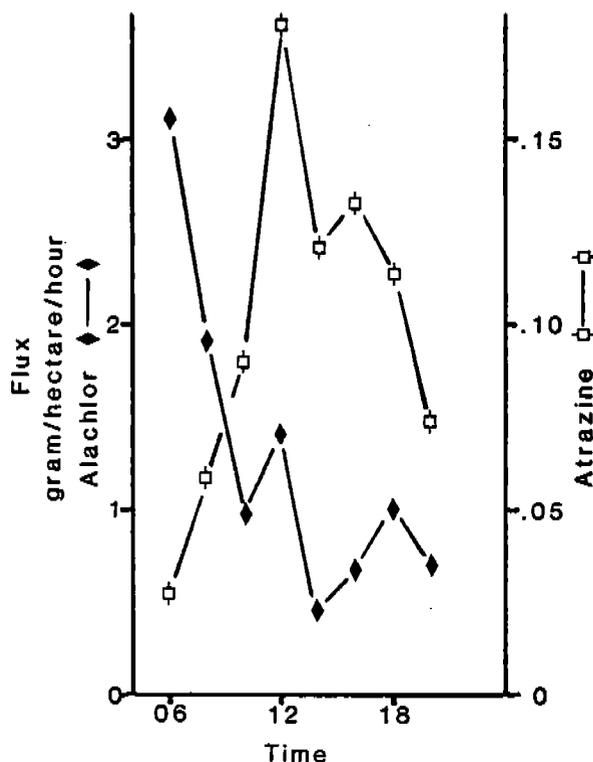


Figure 34.1. Field-measured rates of loss of atrazine and alachlor to the atmosphere on the third day following their application to bare soil.

0.1% per day, which, if sustained, would be comparable under some circumstances to the runoff losses of these chemicals.

We measured atrazine levels in the general atmosphere at several locations across Maryland. We found that even though atrazine has a very low vapor pressure it is present in the atmosphere at all seasons, even in winter, and fairly uniformly distributed over wide areas. From mid-April to mid-July, background air concentrations generally ranged from 2 to 20 ng/m³, with less than 10% trapped in filters. This suggests the possibility that even atrazine may exist in the atmosphere to a significant extent in the vapor phase. In winter, concentrations were much lower, about 0.02 ng/m³, and about half was retained by filters.

As shown in Figure 34.2, atrazine concentrations in rain also change with season. Peak concentrations (about one ppb) occur at local corn-planting time. The maximum deposition rate we observed was about 50 µg/m²/week. (On an area basis, treatment rates to control weeds in corn are 30,000 times greater.) During the winter, precipitation concentrations were fairly constant, averaging about 20 ppt. We did observe a small but significant rise in late January, corresponding to use of the herbicide in other parts of the nation. Evidently, atrazine may travel a considerable distance in the atmosphere.

We would like to be able to predict how effectively precipitation removes pesticides from the atmosphere. Rainout efficiency is determined by comparing

the volatilization (Caro et al. 1976).
 case. Volatilization from soil peaked in the first 6.8% of the runoff; see Caro et al. 1976. Volatilization from the mature corn

or to vegetation, 90% within six to eight periods, both measured and estimated proportional to their surface area. Over vegetation, air flow rates usually become moist soil become the spray.

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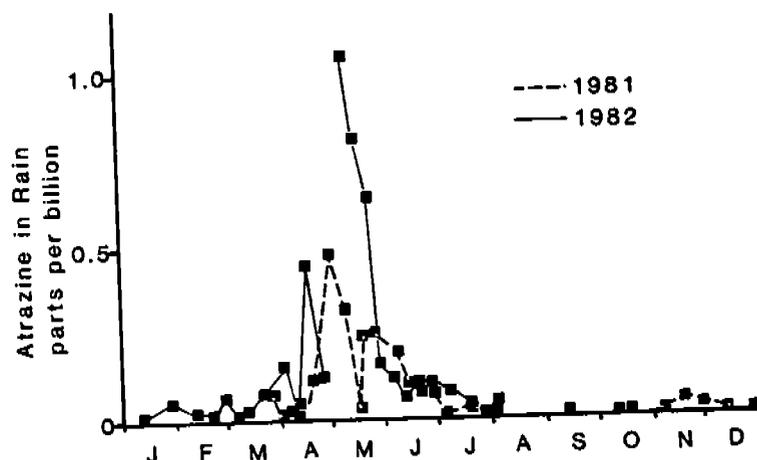


Figure 34.2. Seasonal variation of atrazine concentration in precipitation, measured in Maryland, 1981-82.

the measured relative concentrations in air and rain with the distribution between air and water predicted by Henry's Law coefficients. It may not always be clear what these numbers mean, however. One problem is that we are usually comparing concentrations in ground-level air with those in rain formed at some higher altitude.

Because of its intrinsic properties, atrazine should be readily absorbed from air into rain, yet the rainout efficiencies that we have measured are all low. The evidence suggests, however, that the reverse is true; rain is totally efficient in removing atrazine from that volume of air in which the rain forms. Since this is less than the entire atmosphere, rainout efficiencies appear to be low.

In the early 1970s, the National Air Monitoring Program found that a high percentage of ambient air samples taken throughout the country contained measurable levels of heptachlor (Kutz, Yobs, and Yang, 1976). Yet few of the more recent reports in the open literature, especially those dealing with regional and global transport of organic contaminants, have specifically mentioned heptachlor. It may be that heptachlor sources have dissipated since the 1974 ban, or that heptachlor is too reactive to persist very long in air.

Chlordane, a more stable congener of heptachlor, is dispersed in air on a global scale and is found in very remote, mid-ocean air samples (Atlas and Giam 1981; Bidleman and Olney 1974). Virtually none is retained on filters, and it is thought to exist in the atmosphere almost wholly as vapor. Although we find an average of 45 ppt of chlordane in rain, rain is not as effective in removing chlordane from air as it is in removing other chlorinated compounds (Bidleman, Christensen, and Harder 1981). The primary removal mechanism may be rather slow, direct surface deposition, and chlordane thus appears to have a very long atmospheric residence time.

ENVIRONMENTAL PERSPECTIVES

Intrinsic properties can indicate tendencies for pesticide dispersion. Atrazine and heptachlor have widely different properties and, as expected, atrazine is carried

primarily in water and heptachlor primarily in air. In reality, however, each is carried to a certain degree in both mobile phases.

To successfully model the movement of pesticides to the boundary of the treated field, we must pay proper attention to such details as formulation, application method, and weather. Once outside the treated area the picture becomes less distinct, because the fluid phases, water and air, are extremely heterogeneous and changeable, and there are many ways in which a pesticide may be distributed in the environment.

The formulation used to apply the pesticide can determine transport potential. For example, wettable powder formulations are susceptible to erosion by both water and air, and pesticides are thereby mobilized despite their intrinsic properties. On the other hand, formulations can control losses, as when microencapsulated or granular formulations are used to apply a volatile pesticide (Turner et al. 1978).

How we manage a pesticide also determines movement. One of the worst things we can do, from an environmental perspective, is to spray a pesticide into the air in an aqueous dispersion, because this invariably leads to drift and loss. On the other hand, incorporating a pesticide into soil can greatly reduce its movement in both air and water.

Weather is a factor. For example, how soon rain follows application has much to do with the extent of pesticide movement in runoff. Things are never as homogeneous in the field as we make them in the laboratory, and the profound influence of nonuniform distribution of soil moisture on pesticide volatilization was given as an example of how this was important.

Pesticides may be carried more rapidly and to greater distances by the atmosphere but more quickly dispersed to low concentrations than if the pesticides were in runoff water. Thus, the scale of transport and severity of contamination may depend upon whether the pesticide is carried in water or air, and upon the mixing, residence time, and characteristic chemical reactions within each phase. Pesticides are dispersed on a local and regional scale by water and air, but pesticides currently dispersed on a global scale have properties that allow them to move and survive intact for long periods in the atmosphere.

It is two decades since publication of *Silent Spring* and the introduction of the electron-capture detector for gas chromatography—two landmark events that made us aware of the potential for harm by pesticides and gave us our first good tool to study environmental problems. In the past two decades, analytical capabilities have advanced to the point where truly remarkable sensitivity and selectivity in trace pesticide analysis are possible. With these techniques we can show what should have been clear all along: it is a mathematical certainty, consonant with the laws of thermodynamics, that each time a pesticide is used some form of environmental contamination will result.

Often, our instruments allow us routinely to measure quantities so small that they are difficult to comprehend. Figure 34.3 gives an example, which shows the gas chromatographic analysis and chemical derivative identification of simazine in a rain sample collected in Maryland during a 2-week interval ending 20 January 1982. The top chromatogram is that of the sample; the bottom, reference standards. The analytical procedure required sample collection, extraction, and concentration of the herbicide, fraction of the sample by adsorption chromatography, residue polishing by reversed-phase high-pressure liquid chromatography, and finally, gas chromatographic detection using a detector sensitive only to nitrogen. It is obvious that the chromatogram is clean and the measurement of simazine is unambiguous.

The average simazine concentration in the rain was about 9 parts per trillion

January 20, 1982

Rain Analysis

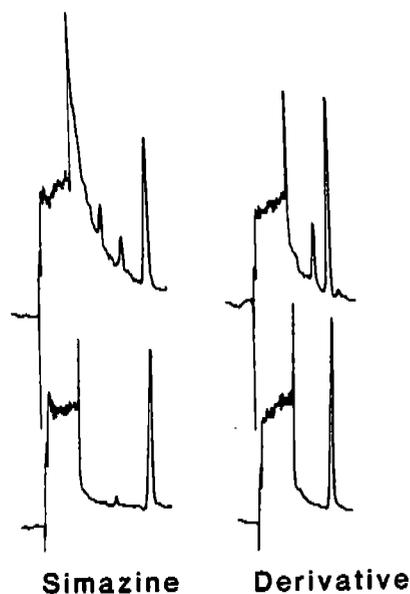


Figure 34.3. Gas chromatographic analysis and chemical derivative (methoxylation) identification of simazine in mid-winter rain collected in Maryland. The top chromatograms are those of the sample; the bottom, reference standards. Simazine concentrations was 9 parts per trillion.

(ppt). The total deposition over the 2-week period was about 160 nanograms/m²—slightly less than 1.5 ounces per 100 square miles! Pesticide concentrations are often expressed as weight ratios; for example, a part per billion (ppb) may mean one gram of pesticide in a billion (10⁹) grams of soil. This is difficult for me to visualize. Since these are ratios of physical quantities, they may be illustrated with ratios of other physical quantities, such as length and time. In terms of time, a part per million (ppm) is one second in 11.6 days; a ppb is one second in 32 years; a ppt is one second in 32,000 years. On this basis, the simazine concentration in rain is roughly equivalent to one second in the time elapsed since three centuries before the Trojan War.

Since they are all man-made, pesticides in the environment are present in excess of their occurrence in the absence of human activity. They are thus, by definition, environmental contaminants (Dunn 1980). A discussion of pathways of environmental dispersion is, of necessity, a description of the natural events that lead to environmental contamination. The evidence suggests that simazine had traveled a long distance in the atmosphere before falling with precipitation, and our analytical prowess clearly proves that simazine is a widespread environmental contaminant, touching many parts of the biosphere. However, because of its chemical and biological properties, such vanishingly small quantities do not poison throughout, and simazine delivered in rain is not an environmental pollutant. We need to be aware that there is a difference between contamination and pollution, and that we are each in part responsible for choices concerning the use of agricultural chemicals. The demand for using these products results as much from consumers' preferences for low-cost, high-quality food as from a desire by farmers to earn a profit.

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