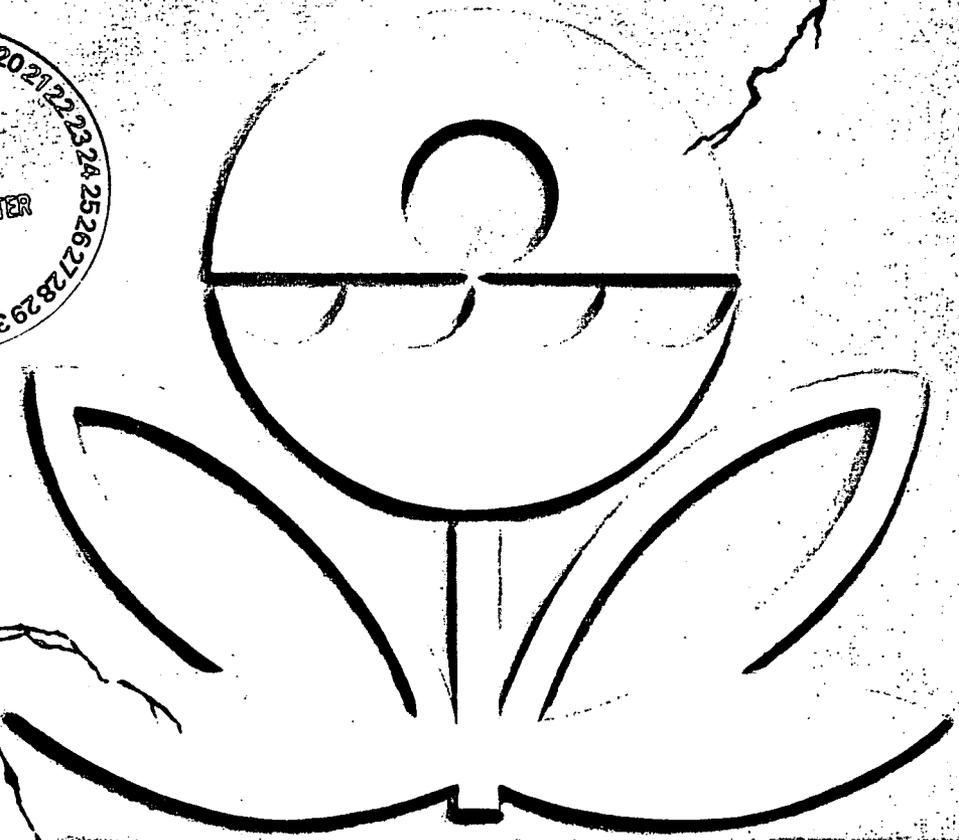


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How Toxic Are Toxic Chemicals in Soil?

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Assessments of the hazard of toxic chemicals in soil are made without concern about the possibility that their bioavailability may change with time. The patterns of disappearance of persistent compounds in the field and laboratory studies show a declining availability to microorganisms with residence time in soil. Changes in extractability with residence time and the kinetics of sorption and desorption suggest that the compounds are becoming sequestered in inaccessible microsites within the soil matrix. Diminishing toxicity as chemicals age in soil is evident in a limited number of assessments. Such findings suggest that the hazard and risk from toxic chemicals diminish as the compounds persist in soil.

Most polluted soils or subsoils that are currently being considered for remediation were contaminated many years ago. These soils were contaminated before there was widespread concern with environmental deterioration and before strict regulations were established and the high cost of remediation became evident. Nevertheless, assessments of the hazards from those sites have not taken into account slow processes that may take place and possibly reduce the impact of toxic compounds deposited in the soil, particularly those compounds that do not leach out to contaminate underlying aquifers. In recent years, evidence has accumulated that the availability of certain organic compounds changes as the compounds reside in soil for some time, a process that has been termed aging. Data have also been collected suggesting that organic molecules slowly become sequestered within the soil matrix. The declining availability and sequestration appear to be related, and a consideration of the declining bioavailability and the occurrence of chemical sequestration has great relevance to assessing toxicity, determining risk, and establishing meaningful regulations for the cleanup of sites containing hazardous wastes.

Three lines of evidence point to the sequestration of organic molecules that persist in soil: (a) field and laboratory studies demonstrating a diminishing availability to microorganisms; (b) investigations of the extractability of aged chemicals and the kinetics of sorption and desorption; and (c) assessments of toxicity. Although the issue of toxicity is the most relevant for decisions on risk and for

environmental regulations, that line of evidence is based on few studies. However, all three lines of evidence are consistent and point to the need for a modified approach to assessing risk.

Diminishing Availability to Microorganisms

Long-term monitoring of organic compounds in soils has been chiefly restricted to pesticides, especially the chlorinated hydrocarbons that were once widely used as insecticides. These field measurements do not distinguish among losses resulting from biodegradation, volatilization, or abiotic decomposition. However, all the pesticides are biodegradable so that a chemical which disappeared initially but not in later periods must be less susceptible to all loss mechanisms, including degradation by microorganisms in the soil. The disappearance of appreciable amounts of these insecticides from the field sites was not a result of leaching because all are extensively sorbed and little vertical movement has been detected even after many years.

These persistent pesticides initially disappear from soil at reasonable rates, but frequently the rate subsequently slows markedly. A typical example is DDT [1,1,1-trichloro-2,2-bis(*p*-chlorophenyl)ethane]. Almost three decades ago, Nash and Woolson (1) reported a slow but continuous disappearance of the compound in the 10 years after its addition, but little or none of the compound was lost in subsequent years (Figure 1). Because this insecticide is metabolized by many microorganisms and microbial activity is a major contributor to its disappearance (2, 3), prevailing conditions were conducive to their activity. However, some slow change occurred with the passage of time that rendered the compound increasingly less available to the microflora. Low winter temperatures and occasional drought periods can be ruled out for the lack of detectable biodegradation because the persistence curve extends for numerous seasons and many years. Similar curves, each with an initial phase of loss followed a period of little or no detectable loss, have been reported for DDT added to many soils in many areas of the world from the 1960s to the present time (3-6).

Field monitoring has shown analogous "hockey-stick" shaped curves for a variety of other chlorinated hydrocarbons; e.g., aldrin and its epoxide, dieldrin (1, 4, 7, 8), heptachlor and its epoxide (1, 9, 10), chlordane (1, 4), kepone (11), nonylphenol, and a linear alkylbenzene sulfonate (12). Examination of the data from these field monitoring

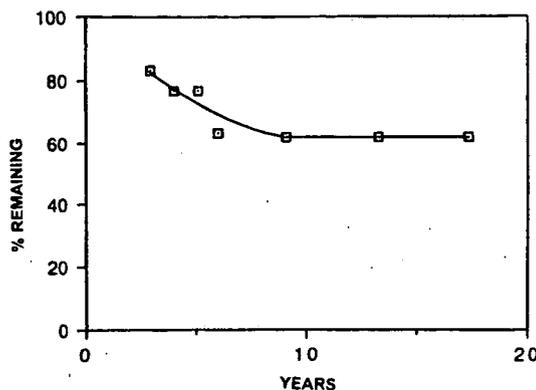


FIGURE 1. Concentration of DDT in Chester loam amended with 200 mg of the insecticide/kg of soil. Replotted from the data of Nash and Woolson (7).

programs reveals enormous differences in the time at which the initial phase of decline ends and the percentage of the original compound that remains following that decline. The time is sometimes as short as 1 year, but it may be longer than 10 years. From 10 to about 60% of the original compound remains in the soil during the period when there is little or no loss. Even with a single compound, the times and percentages may vary greatly; e.g., for DDT, aldrin, and dieldrin. A relationship to soil type or climate is not evident from the available data, but a systematic study was never conducted to establish such a relationship. It is possible that these differences in times and percentages can be attributed to the relative rates of degradation (or loss by volatilization) and the sequestration of the compounds in unavailable forms. If the loss is rapid compared to sequestration, little will remain. If the loss is slow and sequestration is rapid, a higher percentage will remain. Moreover, if the loss is so rapid that little time elapses for the slow sequestration, a residual, persistent amount will not be detected; this is the case for the many pesticides and other organic compounds that rapidly disappear from soil because of their degradation, volatilization, or leaching.

Laboratory studies confirm the unavailability to microorganisms of molecules that have been in soil for long periods. Thus, little or no loss was detected of 1,2-dibromoethane in a soil treated in the field 3 years earlier and of simazine [2-chloro-4,6-bis(ethylamino)-s-triazine] from a field that received the herbicide for 20 consecutive years, although newly added ^{14}C -labeled 1,2-dibromoethane and simazine were extensively metabolized in several weeks (13, 14). Similarly, polycyclic aromatic hydrocarbons present in a soil at a manufactured gas site did not disappear in a 3-month period in the laboratory, but naphthalene and phenanthrene freshly added to the soil were rapidly metabolized (15).

A somewhat different approach to demonstrate aging was used by Hatzinger and Alexander (16). They introduced phenanthrene, a hydrocarbon that is not readily lost by abiotic mechanisms, into sterile soil and added a phenanthrene-degrading bacterium after the hydrocarbon had aged for different periods of time. The data with a soil rich in organic matter show that the extent of microbial conversion of phenanthrene to CO_2 and the rate of biodegradation decline with increasing time of contact with the soil (Figure 2). Similar observations were made with a soil with a lower level of organic matter, although aging appeared to be slower.

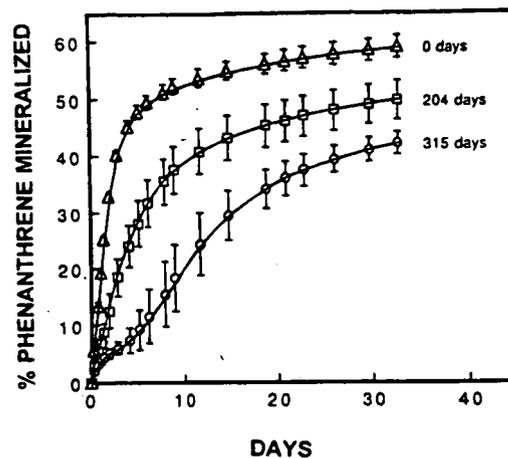


FIGURE 2. Biodegradation by a strain of *Pseudomonas* of phenanthrene at 10 mg/kg that had aged for 0, 204, or 315 days in Edwards muck. The error bars represent the standard deviations (16).

These field and laboratory investigations show that some abiotic process(es) make(s) organic compounds less readily available to microorganisms. The impact of that process or those processes becomes greater as the residence time of the chemicals increases. The fact that such a change in bioavailability affects microorganisms is of relevance to higher organisms because presumably bacteria are more able than plants and animals to assimilate chemicals in soil in view of their small size, their high population densities, and the physiological versatility of the indigenous microflora.

Extractability, Sorption, and Desorption

Little difficulty is usually encountered in finding an extractant that gives quantitative recoveries of organic compounds shortly after their introduction into soil. However, if a chemical persists and thus remains in contact with the particulate matter for some time, it becomes more and more resistant to extraction by many solvents. This has been known for many years by chemists endeavoring to obtain quantitative recoveries of insecticides and herbicides used for pest control (17-19). A diminished availability for solvent removal also has been noted in a sterile organic matter-rich soil and a loam that was incubated with phenanthrene under aseptic conditions (16). The organic solvents that extracted all of the newly added compound removed increasingly smaller amounts as the chemical persisted. More recent studies show that phenanthrene aged in sterile soil similarly becomes, with time, progressively less extractable with acetonitrile/water (1:1) and various ethanol/water mixtures (B. D. Kottler, J. W. Kelsey and M. Alexander, unpublished data). This represents another line of evidence that a change is occurring that renders persistent molecules less accessible, whether accessibility is assessed in microbiological or chemical terms.

Molecules that behave in this way are not to be confused with bound residues. Such bound residues, which have been well studied for pesticides, are those residual fractions that are not extracted by procedures that do not appreciably alter the nature of those residues; these molecules are changed in some manner and are usually converted to the original compound by vigorous hydrolysis. In contrast, aged compounds can be extracted by some organic solvents, often under vigorous conditions, and are thus subject to regulation if the original compounds are toxic in solution.

Studies of kinetics of sorption and desorption provide another perspective on the behavior of toxicants that is relevant to issues of risk. For hydrophobic as well as some other compounds, the attainment of apparent equilibrium in sorption may require weeks, months, and sometimes possibly even years; i.e., more and more of the organic compound is sorbed by soil particles with the passage of time. The initial phase of sorption is rapid, and frequently about half the chemical in aqueous solution is removed by the soil in a few minutes or hours. This first phase is followed by a considerably slower uptake, which can be of prolonged duration. Such kinetics have been observed not only with soil but also with aquifer solids (20-24), and they suggest that the sorption involves not only the external surface of the particles but also a slow and continuing diffusion of the molecules to sites within the particles. The internal and more remote sites continue to bind more of the compound with increasing time.

The desorption of many chemicals similarly shows a rapid phase followed by a period of slow desorption. Such behavior has been noted for polychlorinated biphenyls, trichloroethylene, tetrachloroethylene, toluene, xylene, picloram (4-amino-3,5,6-trichloropicolinic acid), and atrazine (2-chloro-4-ethylamino-6-isopropylamino-s-triazine). Furthermore, the longer the compound remains in soil, the less is the amount released (24-27). Such data suggest that, following the initially rapid release from sites very near to the particle-water interface, the slow appearance of the chemical in solution results from molecules originating from sites at some distance from the interface. The longer aging time allows more of the molecules to diffuse to ever more remote sites. Furthermore, the fraction of a compound that is not readily desorbed under natural conditions will increase with time because that is the fraction that will be less readily biodegraded, leached, or volatilized.

Sorption of nonionic compounds in soil is viewed as a three-dimensional process. The longer a chemical remains in the soil, the greater is the amount present in remote sites and the further is the bulk of the molecules from the more accessible, outer sites (28). Aging thus is presumably associated with a continuous diffusion into more remote sites, where the molecules are retained. Such a conceptualization is consistent with the findings of initially rapid followed by slow phases of both sorption and desorption. Furthermore, this view may explain the decreasing availability to microorganisms of organic compounds as they undergo aging, and it is of direct relevancy to the issue of toxicity to higher organisms of chemicals that have been in soil for extended periods.

Intraparticle or intraorganic matter diffusion is believed to account for the slow phases of sorption and desorption of nonionic molecules. According to the intraparticle diffusion model, the solute is within micropores inside of soil particles. Diffusion is greatly retarded by (a) the partitioning of the compound between the liquid in the pore and the pore wall and (b) the tortuous path between micropores before the compound reaches the outer surface of the micropore-filled soil particle (29, 30). Although the diameters of micropores within soil particles extend over a wide range, a considerable part of the pore volume is made up of pores with effective diameters of $<1.0 \mu\text{m}$, and examination of several soils and solids from aquifers reveals an abundance of pores with diameters of 20 nm or smaller (31-34). Such pores are so small that even the smallest

bacterium, animal, root, or root hair could not penetrate.

Intraorganic matter diffusion of hydrophobic molecules may essentially reflect a partitioning into the native organic matter of soil, the molecules moving into the amorphous humic polymers in a fashion similar to the movement of a hydrophobic compound from water to an organic solvent. However, the importance of partitioning into soil organic matter to slow sorption and desorption has been questioned (35, 36). Nevertheless, intraorganic matter diffusion is cited as being important for the slow sorption of a number of hydrophobic compounds by soil (35).

Regardless of which mechanism applies to a particular site or which is more important for a given soil, the outcome for bioavailability and toxicity of molecules is, for all intents and purposes, the same. Whether the molecule is present in a remote micropore, has partitioned into some solid organic phase in the soil, or both, it has become sequestered. In this sequestered state, it is inaccessible to microorganisms, plants, and animals. This physical remoteness of aged compounds and their very slow diffusion to locations that are biologically inaccessible are key considerations in assessing risk of toxicants in soil.

Assessments of Toxicity

The line of argumentation presented above—which follows from knowledge of the chemistry of sorption and the effect of aging on the availability of biodegradable substrates to microorganisms—is scarcely reflected in the literature of toxicology. Indeed, not a single investigation has been conducted of the possibly diminishing effect on mammals as toxicants age in soil, although in several studies, samples were taken from soils that were contaminated long before the assay was conducted. The difficulty in interpreting these investigations in terms of an aging effect is the inability to determine whether the diminished toxicity occurred only immediately after the compound came into contact with soil or whether there was an increased diminution of toxicity as the compound became sequestered in soil. Many toxicants become less hazardous to test mammals within a short time after they are added to soil or aquifer solids, regardless of whether exposure is by oral or dermal routes; e.g., trichloroethylene, benzo[a]pyrene, 2,3,7,8-tetrachlorodibenzo-p-dioxin (TCDD), and *m*-xylene (37-40).

Such reservations on the possible involvement of aging must thus be considered in interpreting data showing that compounds present in soil for long periods are less toxic than the same compounds provided to test animals in the absence of soil. For example, less TCDD was taken up by rabbits and guinea pigs and far fewer of the guinea pigs were killed following their oral exposure to soils that had been contaminated years earlier than following exposure to newly treated soils (41, 42). A surprising effect of short-term aging was noted by Poiger and Schlatter (40), who found that less TCDD was absorbed by mammals from soil that had been in contact with this dioxin for 8 days than for 10-15 h. The work of Edwards et al. (43), although with insects rather than mammals, is noteworthy. Chemical determination and quantitative bioassays of the toxicity of lindane (γ -isomer of hexachlorocyclohexane) to *Drosophila melanogaster* were in good agreement shortly after adding this insecticide to the soil, but the results of the bioassays suggested that less of the toxicant was present after 22 months than shown by chemical analysis; i.e., although all of the compound was initially bioavailable, only a part was affecting the fruit flies in soil in which the molecule had

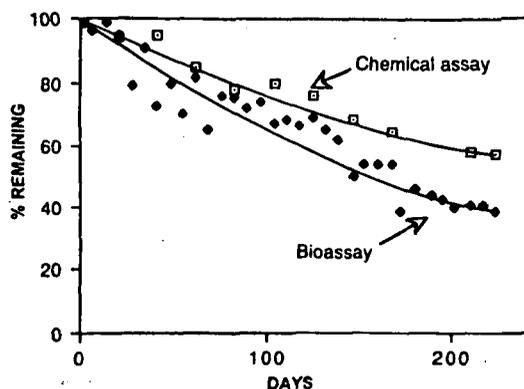


FIGURE 3. Changes with time in concentration of napropamide in soil as measured by bioassay and by extraction followed by gas chromatography. Calculated from the data of Hurle (45).

persisted for almost 2 years. An effect of aging also is suggested by a report that sugarbeet (*Beta vulgaris*) seedlings were not inhibited when grown in a fine loam that contained 0.22 mg of simazine/kg of soil derived from a field that was amended with the herbicide for 20 consecutive years, although 26 of 51 sugarbeet seedlings when grown in the same soil freshly amended with simazine at 0.22 mg/kg were injured by the newly introduced chemical (14).

Additional evidence that some compounds become progressively less toxic with increasing residence time comes from several old and largely forgotten studies. In one, the toxicity of DDT was found to decrease with an increase in the time during which the compound remained in soil (44). In another study, the rate of disappearance of napropamide [2-(α -naphthoxy)-*N,N*-diethylpropionamide] was faster when measured by quantitative bioassays of toxicity of the herbicide-treated soil to oats (*Avena sativa*) than when determined by gas chromatography (Figure 3); i.e., decreasing percentages of the herbicide were toxic with time (45). Similarly, bioassays with *Sinapis arvensis* (mustard) of soil to which was applied chlorbromuron [1-(3-chloro-4-bromophenyl)-3-methoxy-3-methylurea] gave higher values at approximately 30 days, the same values at approximately 60 days, and lower values at 85, 110, and 140 days, thereby showing that not all of the herbicide was available for toxicity (46). Aging also appears to occur in lake sediments, as witnessed by the decline in the rate of uptake of pyrene by the amphipod *Diporeia* sp. (47). Furthermore, we have recently found that phenanthrene aged for up to 150 days in samples of two sterile soils becomes progressively less available to earthworms subsequently added to these soils (J. W. Kelsey and M. Alexander, unpublished data).

A diminishing or reduced bioavailability with time is not always evident. The absence of an aging effect has been noted in investigations of the dermal availability to rats of TCDD (48), the toxicity of some herbicides to plants (46), and the biodegradation of a number of persistent pesticides by microorganisms (14). The reasons for the absence of an effect are as yet unknown, but they may be related to such properties of the soil as organic matter content or abundance of micropores or to such properties of the compound as hydrophobicity.

Perspective

The information that has been presented shows that (a) the availability of long-lived organic chemicals to microorganisms in soil in the field declines markedly with time,

(b) some freshly added chemicals are readily available to microorganisms in soil in which the identical but aged compounds are not metabolized by indigenous microorganisms, (c) organic compounds incubated in sterile soil become increasingly less available to subsequently added microorganisms, (d) some compounds become increasingly resistant with time to extraction, and (e) sorption and desorption of hydrophobic compounds often require long time periods to reach equilibrium. These five lines of evidence are consistent with a sequestration of the molecules within particulate matter, presumably because the molecules diffuse into internal micropores or sites that are spatially remote. Because of their unavailability to even minute organisms and many extractants, it is likely that the compounds are also unavailable to humans, animals, and plants. These five lines of evidence are also consistent with a small but growing body of information showing that bioremediation of hydrocarbon-contaminated soils markedly reduces their toxicity to test species, despite the presence of presumably toxic concentrations of hydrocarbons as determined by exhaustive extraction (49).

Such a conclusion in regard to sequestration has major implications for assessments of the toxicity of compounds in soil that are known to be harmful in the absence of soil. The small base of information cited above shows that, at least for a few organic compounds, the toxicity to animals and plants disappears faster than the chemical itself. Thus, sequestration appears to alter the hazard to higher organisms. Indeed, the current views of the processes underlying slow sorption and desorption and those that give rise to ever increasing resistance to extraction implicitly suggest that toxic molecules will be less hazardous because they move to internal sites in soil particles that are too remote to impact living organisms.

The processes occurring may be envisioned in the following fashion. A toxic compound in solution is initially sorbed rapidly to the external surfaces of soil. This fraction is available for rapid desorption, to many organic solvents, to microorganisms for biodegradation, and to have some detrimental effect on susceptible organisms. That available fraction is slowly converted to an unavailable fraction, which is remote from the external surface. This portion of the compound is essentially irreversibly sorbed, is only extracted by highly vigorous means, and is neither available to microorganisms nor to cause injury to susceptible humans, animals, and plants. Not all of the compound will be sequestered and become nontoxic because soils into which 2,3,7,8-TCDD, polychlorinated biphenyls, or polybrominated biphenyls had been introduced years earlier still have considerable mammalian toxicity (41, 42, 48, 50-52). This toxicity is unlikely to have resulted from chemicals in solution because each of those compounds is extensively sorbed.

From the information presented, it is clear that regulatory decisions based on the currently used vigorous extraction procedures, which are designed to remove as much of the toxicant from soil as possible, may overestimate risk. This is not surprising, not only in view of the information cited above but also from an extensive literature that shows that the toxicity of the same concentration of an insecticide or herbicide may be vastly different in soils with dissimilar properties (43, 44, 53, 54).

Because of the sequestration of toxicants in soil, evaluations need to be made of the possibly reduced risk to humans, animal populations, plant populations, and

ecosystems. The very fact that the organic compounds in Superfund and most other hazardous waste sites have been in the contaminated soils for long periods of time emphasizes the need for assessing the significance of aging to toxicity. Tests of toxicity with pure chemicals in the absence of soil or chemicals freshly added to soil probably overestimate risk. Exhaustive extraction for determining toxicants in soil may be unreliable for assessing the need for and effectiveness of remediation. An approach more meaningful than the ones used presently to establish priorities for remediation would include an assessment of the degree to which sequestering in soil alters the availability of environmental pollutants.

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