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PLANT UPTAKE OF NON-IONIC ORGANIC CHEMICALS FROM SOILS

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ABSTRACT

Methodologies utilizing simple properties of chemicals - half-life ($T_{1/2}$), log octanol-water partition coefficient (log K_{ow}) and Henry's Law constant (Hc) - are developed to screen organic chemicals for potential plant uptake.

INTRODUCTION

Early in 1983, the American Chemical Society's Chemical Abstract Service registered its 6,000,000th chemical. The Toxic Substance Control Act Inventory list 63,000 chemical substances whose manufacture, processing and ultimate use for commercial purposes has occurred in the United States since January, 1975 (TSCA Inventory, USEPA, 1985). Additionally, the number of synthetic organic chemicals used and disposed of by society is increasing at a rate of about 1000 new chemicals per year, (Loehr and Malina, 1986). This endless supply of compounds together with the variety of reactions they can undergo in the environment makes describing their environmental impact exceptionally challenging.

Of the possible locations for the disposal of wastes - surface waters, atmosphere or land the latter represents a common location for waste disposal as well as an opportunity to manage wastes with minimal environmental impact. The object of the land disposal practice is to degrade, immobilize, and/or transform the wastes into beneficial, or at least non detrimental constituents. There are over 200 industrial waste land treatment sites in the United States, and a larger number of land treatment sites for municipal wastewater and sludge (Loehr and Malina, 1986). Land disposal of wastes has increased during the past decade and is projected to continue to increase in the future (Loehr and Malina, 1986).

The study of organic chemicals in the soil environment has been dominated by agricultural chemicals (e.g., insecticides, nematicides and herbicides) and specific compounds that persist in the soil (e.g., PCB's, PBB's etc.). This narrow perspective probably occurred because of the prevalence of agricultural chemicals in soil, complexity of reactions, large number of compounds, and cost associated with organic analysis. Specific compound attention has been propagated by the formation of lists of specific compounds, such as the organic priority pollutant list of 1976. Even with this narrowing of focus, the cost associated with a chemical by chemical investigation is prohibitive. The approach therefore

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has been to utilize physicochemical parameters, or to group compounds on the basis of their chemical or physical properties and study selected compounds from each group. Clearly, we must insure that the grouping of compounds is correct and that the factors used in the groupings predict the behavior and impact of compounds not studied.

The following attempts to provide a framework which uses physicochemical parameters to evaluate potential plant uptake of neutral or weakly ionized organic chemicals from soil. The procedure does not predict plant concentration of organics in a field situation, but provides a procedure for grouping chemicals by their relative potential for plant uptake. As such, it should allow compound screening for their likelihood for plant uptake and, therefore, justify experimental evaluation as well as identify chemicals of low concern where testing may be counterproductive. It should also reveal where information is needed to confirm the screening model.

BEHAVIOR OF ORGANIC CHEMICALS

Many processes impact organic chemicals in the soil environment. The sum of these actions determine the compounds environmental impact (Figure 1). Factors such as pH, CEC, OM content, clay content and soil water content all impact the rate and extent of these processes (Goring and Hamaker, 1972). In a given situation (soil and environmental conditions) however, the processes are dependant upon the physical and chemical properties of the chemical. The characteristics of a chemical that determine its distribution between vapor, solid, liquid and adsorbed phases in the soil, and its degradation rate become the characteristics that determine its environmental fate and impact upon plants. These processes determine not only the form of the compound that is present, but also the speed at which the compound moves or spreads through the soil and atmosphere to achieve its impact. The importance of each of these processes will be discussed separately.

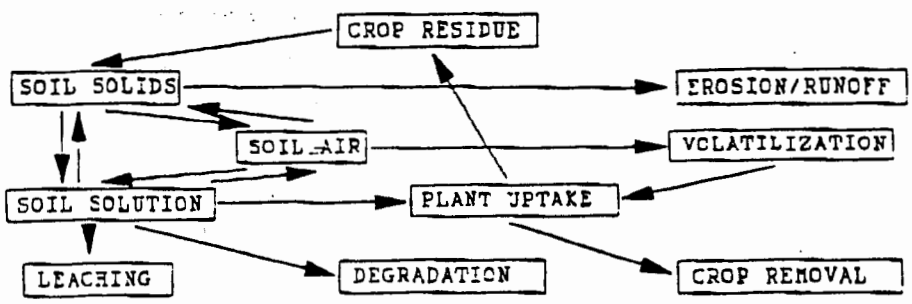

FIGURE 1 SOIL TRANSFORMATIONS

Degradation

Plant uptake of most chemicals is concentration dependent, therefore a compound's persistence can alter its ultimate fate and environmental impact. An assessment of the half-life of a particular compound is a relatively simple way of limiting the number of soil borne organic compounds that need to be considered as likely to impact a plant grown in contaminated soil. The concentration of synthetic organic compounds in the soil decrease with time, providing no further additions occur. Processes contributing to the decrease with time are biological and/or chemical degradation. These processes have been shown to be dependent on soil and environmental factors (ie., temperature, water content, soil pH, and organic C). (Hamaker, 1972). Without the quantitative information necessary to describe the functional dependence of degradation on these factors, it has been shown that degradation of a specific organic chemical can be described by a first order rate constant, μ, (Nash, 1980; Rao and Davidson, 1980; Jury et al., 1983; Gillett,1983). This parameter is usually measured by determining the fraction of an applied chemical remaining after a time t according to Equation 1 :

$$M(t) = M(0) \exp^{(-\mu t)} \qquad [1]$$

where M(t) is the quantity of the compound remaining in the soil at time t. The half-life, $T_{1/2}$, of a compound is defined as the time required for one half of the concentration of the chemical at any point in time to be lost from the soil. This is related to the rate constant (μ) by :

$$T_{1/2} = \frac{0.693}{\mu} \qquad [2]$$

Half-lives of many chemicals have been published (USEPA, 1979; Jury et al., 1983; Smith and Dragun, 1984). Unfortunately, reported values of μ may vary enormously because measured half-lives of compounds in the soil do not always reflect degradation. Often losses include other pathways (i.e., volatilization, leaching, etc.). Additionally, water content, microbial population, and temperature can significantly influence the rate of loss thus, a chemicals life may vary from soil to soil. Half-lives are reported in Table 1 from data in USEPA, 1979. Compounds are distinguish from one another on the basis of half-life in the soil: less than 10 days, (Class A); between 10 and 50 days, (Class B); and greater than 50 days, (Class C). Gillett considered compounds of $T_{1/2}$ greater than 14 days of sufficient stability to be of concern (Gillett, 1983). The impact of chemical half-lives on concentration of a pollutant in the soil over time is shown in Figure 2. Pollutants with half-lives of less than 10 days, for example, are reduced to less than 0.10% of their original concentration after 100 days in the soil. In contrast, pollutants with half-lives of greater than 50 days are still present at >25% of their original concentrations after 100 days. Their impact, and relative potential for plant uptake, are much more pronounced than that for

compounds with half lives of less than 10 days.

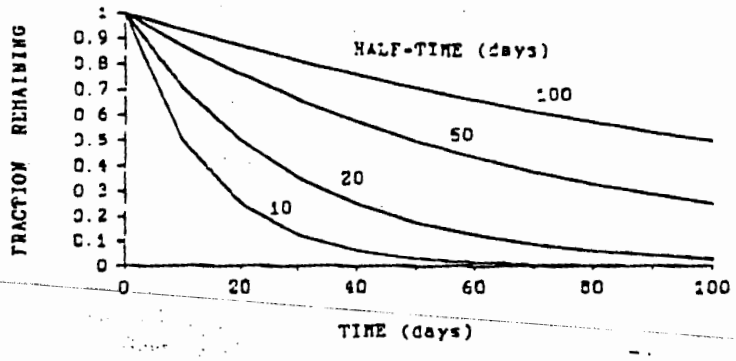

FIGURE 2 EFFECT OF CHEMICAL HALF LIFE AND TIME ON FRACTION REMAINING

The average concentration present during the plant growing period can be calculated by integration of Equation 1 between the limits 0 and t (growth period) and dividing by t. Assuming a growth period (i.e. 50 or 100 days) the effect of half-life on the average soil concentration as a fraction of the amount originally applied illustrates that the limits for classification of compounds based on half-lives are arbitrary (Figure 3). The length of exposure (i.e. plant growth period) and relative average exposure must be specified before compounds can be classified by their half-lives. For example, our use of 10 and 50 day half lives as classification end points was based on a 100 day growth period and relative average exposures of 0.15 and 0.5. Using the same half-life end points but a 50 day growth period means relative average exposures of 0.3 and 0.7.

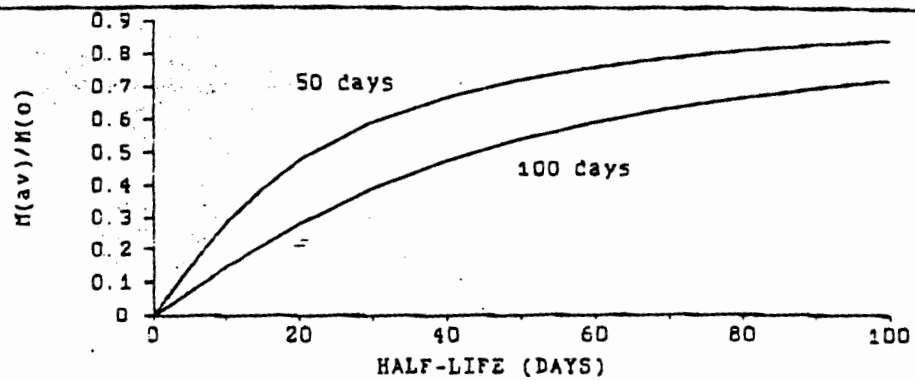

FIGURE 3 AVERAGE SOIL CONCENTRATION VS HALF LIFE FOR 50 AND 100 DAYS OF GROWTH

TABLE 1. Log K_{ow}, Half-life and Hc for the Priority Pollutants

Compound	log K_{ow}	$T_{1/2}$	Hc	Compound	log K_{ow}	$T_{1/2}$	Hc
PESTICIDES							
20.Acrolein	-0.09	B	2.8E-03	21.Aldrin	4.62	C	4.9E-06
22.Chlordane	4.3	C	3.9E-03	23.DDD	5.99	C	0.5E+00
24.DDE	5.69	B	9.0E-06	25.DDT	5.98	C	2.0E-03
26.Dieldrin	2.9	C	3.0E-04	27.Endosulfan	3.55	C	nd
28.Endrin	4.5	C	1.7E-05	29.Heptachlor	3.9	A	6.2E-02
30.Heptachlor epoxide	3.9	C	3.2E-05	31.Hexachlorocyclohexane	3.8	B	3.0E-06
32.Lindane	3.72	C	6.0E-06	33.Isophorone	1.70	nd	nd
34.TCDD	6.14	C	nd	35.Toxaphene	3.85	C	2.1E-01
POLYCHLORINATED BIPHENYLS							
36a.Arochlor 1016	4.38	C	8.6E-01	36b.Arochlor 1221	4.09	C	1.3E-02
36c.Arochlor 1232	4.54	C	2.1E+00	36d.Arochlor 1242	4.11	C	5.5E-02
36e.Arochlor 1248	5.6	C	1.1E-01	36f.Arochlor 1254	6.04	C	1.1E-01
36g.Arochlor 1260	6.11	C	2.9E-01	37.2-chloronaphthalene	4.12	C	1.3E-02
HALOGENATED ALIPHATIC HYDROCARBONS							
38.Chloromethane	0.91	C	1.6E+01	39.Dichloromethane	1.25	B	8.5E-02
40.Trichloromethane	1.9	B	1.2E-01	41.Tetrachloromethane	2.64	nd	9.6E-01
42.Chloroethane	1.54	B	6.1E-01	43.1,1-dichloroethane	1.79	B	1.7E-01
44.1,2-dichloroethane	1.48	B	3.8E-02	45.1,1,1-trichloroethane	2.17	nd	1.3E+00
46.1,1,2-trichloroethane	2.17	nd	3.1E-01	47.1,1,2,2-tetrachloroethane	2.54	A	1.6E-02
48.Hexachloroethane	4.62	nd	4.1E-01	49.Chloroethene	0.60	A	6.2E-01
50.1,1-dichloroethene	1.48	A	1.8E-01	51.1,2-trans-dichloroethene	1.48	A	2.7E-01
52.Trichloroethene	2.29	A	3.8E-01	53.Tetrachloroethene	2.88	A	6.4E-01
54.1,2-dichloropropane	2.28	nd	1.2E-01	55.1,3-dichloropropene	1.98	A	5.6E-02
56.Hexachlorobutadiene	3.74	C	4.3E-01	57.Hexachlorocyclopentadiene	3.99	A	1.5E+00
58.Bromomethane	1.10	B	4.4E+00	59.Bromodichloromethane	1.88	nd	nd
60.Dibromochloromethane	2.09	nd	nd	61.Tribromomethane	2.30	nd	2.4E-02
62.Dichlorodifluoromethane	2.16	C	6.3E+01	63.Trichlorofluoromethane	2.53	nd	2.4E+00
HALOGENATED ETHERS							
64.Bis(chloromethyl)ether	-0.38	A	8.6E-02	65.Bis(2-chloroethyl)ether	1.58	nd	4.7E-05
66.Bis(2-chloroisopropyl)ether	2.58	nd	4.7E-02	67.2-chloroethyl vinyl ether	1.28	A	1.0E-02
68.4-chlorophenyl phenylether	4.08	nd	1.0E-02	69.4-bromophenyl phenyl ether	4.28	nd	nd
70.Bis(2-chloroethoxy)methane	1.26	C	1.1E-05				
MONOCYCLIC AROMATICS							
71.Benzene	2.13	A	2.3E-01	72.Chlorobenzene	2.84	nd	1.5E-01
73.1,2-dichlorobenzene	3.38	nd	1.5E-01	74.1,3-dichlorobenzene	3.55	nd	1.1E-01
75.1,4-dichlorobenzene	3.55	nd	9.9E-02	76.1,2,4-trichlorobenzene	4.26	A	9.6E-01
77.Hexachlorobenzene	6.18	C	7.0E-02	78.Ethylbenzene	3.15	A	2.6E-01
79.Nitrobenzene	1.85	C	5.4E-06	80.Toluene	2.69	A	2.6E-01
81.2,4-dinitrotoluene	2.01	nd	1.3E-02	82.2,6-dinitrotoluene	2.05	nd	1.3E-02
83.Phenol	1.46	A	1.9E-05	84.2-chlorophenol	2.17	nd	1.9E-06
85.2,4-dichlorophenol	3.3	A	2.3E-06	86.2,4,6-trichlorophenol	3.61	nd	1.7E-06
87.Pentachlorophenol	5.01	A	1.2E-06	88.2-nitrophenol	1.76	C	3.2E-06
89.4-nitrophenol	1.91	B	2.6E-06	90.2,4-dinitrophenol	1.53	C	2.7E-08
91.2,4-dimethylphenol	2.50	nd	7.5E-06	92.p-chloro-m-cresol	3.11	nd	1.0E-06
93.4,6-dinitro-o-cresol	2.85	nd	nd				
PHTHALATE ESTERS							
94a.Dimethyl phthalate esters	2.12	B	8.8E-06	94b.Diethyl	3.22	B	1.9E-03
94c.Di-n-butyl	5.20	B	1.2E-05	94d.Di-n-octyl	9.20	B	1.2E-05
94e.Bis(2-ethylhexyl)	8.73	B	1.2E-05	94f.Butyl benzyl	5.80	B	4.2E-05
POLYCYCLIC AROMATIC HYDROCARBONS							
95a.Acenaphthene	4.13	C	1.0E-02	95b.Acenaphthylene	4.07	C	4.8E-03
95c.Fluorene	4.18	C	4.8E-03	95d.Naphthalene	3.37	C	2.0E-02
96a.Anthracene	4.45	C	1.1E-02	96b.Fluoranthene	5.33	C	6.8E-06
96c.Phenanthrene	4.46	C	1.6E-03	97a.Benzo(a)anthracene	5.61	C	4.1E-05
97b.Benzo(k)fluoranthene	6.57	nd	nd	97c.Benzo(b)fluoranthene	6.06	C	nd
97d.Chrysene	5.61	C	8.8E-02	97e.Pyrene	5.32	C	2.8E-01
98a.Benzo(ghi)perylene	7.23	C	nd	98b.Benzo(a)pyrene	6.04	C	4.9E-01
98c.Dibenzo(a)anthracene	5.97	C	nd	98d.Indeno(123-cd)pyrene	7.66	C	nd
MISCELLANEOUS COMPOUNDS							
99.Dimethyl nitrosamine	0.06	nd	nd	100.Diphenyl nitrosamine	2.57	nd	nd
101.Di-n-propyl nitrosamine	1.31	nd	nd	102.Benzidine	1.81	A	nd
103.3,3-dichlorobenzidine	3.02	A	nd	104.1,2-diphenylhydrazine	3.03	nd	nd
105.Acrylonitrile	0.25	A	3.8E-03				

where log K_{ow} and half-lives (assessed as A = < 10 days, B = 10-50 days, C = > 50 days) are based on the principal fate process in the environment (USEPA,79); where Henry's Constant (dimensionless) has been obtained from the WERL Treatability Database

Adsorbed-Liquid Partition

Considerable research data exists on the equilibrium between an organic sorbed to the soil and that in the soil-water phase. For simplicity, this is often expressed as a linear sorption isotherm (Karickoff, 1981):

$$C_s = Kd\ C_l \qquad [3]$$

where C_s is the sorbed concentration (g/kg soil), C_l is the solution concentration (g/m^3 soil solution) and Kd (m^3/kg) is the slope of the sorption isotherm or distribution coefficient (Kay and Elrick, 1967). Equation 3 assumes complete reversibility and equilibrium between the two phases, which may not strictly occur for some chemicals. Di Toro and Horzempa (1982), reported that the sorptive process of 2,4,5,2',4',5'- hexachlorobiphenyl consisted of both reversible and strongly bound components. Such bound residues could not be extracted by normal analytical techniques, but could be detected by radiolabelling. Similar findings have been reported by others working with herbicides and chlorobenzenes (Khan, 1982; and Scheunert, et al., 1985) and may require the above mathematical approach for sorption be modified to account for bound residuals.

In soils and sediments, where the clay content is relatively low, pollutant sorption occurs primarily on the organic fraction of the soil, (Hamaker and Thompson, 1972; Rao and Davidson, 1980). The degree of sorption of the non ionic organic pollutant is then dependant upon the organic carbon content in the soil, or sediment. Variation between materials, which otherwise exhibit a wide range of physicochemical properties, can then be reduced by defining an organic carbon distribution coefficient (K_{oc}):

$$K_{oc} = \frac{Kd}{f_{oc}} \qquad [4]$$

where Kd is the slope of the sorption isotherm in m^3/kg, and f_{oc} is the organic carbon fraction in the soil or sediment, (Means, et al., 1982). This assumes that all organic matter has the same chemical structure.

K_{ow} is defined as the ratio of the organic chemical concentration in octanol to that in water, when an aqueous solution of the organic chemical is mixed with n-octanol and then the organic chemical allowed to partition between the two phases (Dawson, et al., 1980). There have been many investigations into the relationship between K_{oc} and K_{ow}. Briggs (1973) for example reported:

$$\log K_{oc} = 0.524 \log K_{ow} + 0.62 \qquad [5]$$

from his work with 4 agricultural soils and 30 chemicals chosen for their wide range of properties. Similar relationships, see Equation's 6, 7, 8, 9, and 10, have been reported (Means, et al., 1982; Schwarzenbach and Westall, 1981; Rao, et al., 1982; Karickhoff, 1981; and Brown and Flagg, 1981 respectively).

$$\log K_{oc} = \log K_{ow} - 0.317 \qquad [6]$$

$$\log K_{oc} = 0.721 \log K_{ow} + 0.49 \qquad [7]$$

$$\log K_{oc} = 1.029 \log K_{ow} - 0.18 \quad [8]$$

$$\log K_{oc} = 0.989 \log K_{ow} - 0.346 \quad [9]$$

$$\log K_{oc} = 0.937 \log K_{ow} - 0.006 \quad [10]$$

The relationships are surprisingly similar to one another considering they cover over 100 chemicals, as well as a large number of soils and sediments (Figure 4). Thus when the sorption value of a particular pollutant in a particular soil is not available, advantage can be taken of the relationship between the organic carbon distribution coefficient (K_{oc}) and the octanol water partition coefficient (K_{ow}) of the chemical. Recently, a nonempirical measurement (first-order molecular connective indexes) calculated from the non-hydrogen part of the molecule has been shown to predict the K_{ow} of organic compounds with great success (Sabljic, 1987). As these calculated values for various organic compounds become available it will allow for their use in place of K_{ow} or K_{oc}.

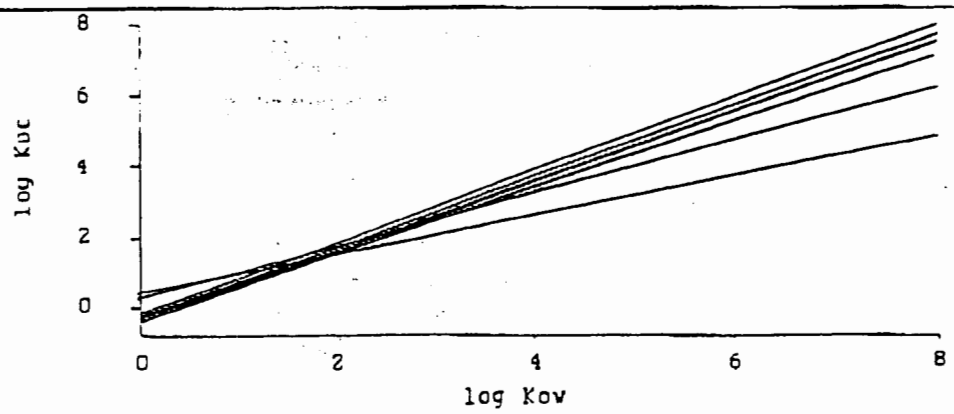

FIGURE 4 RELATIONSHIP BETWEEN log Koc AND log Kow

To have greatest impact upon plant uptake, the organic compound must stay within the vicinity of the plant root, and not be quickly leached away by mass flow. For example most residual soil-acting herbicides have Kd values in the range of 1-20 with values up to 40 being satisfactory for most soil applications (Graham-Bryce, 1984). Compounds with Kd's of greater than 1000 become inactivated by soil sorption (Graham-Bryce, 1984). Based on Equation 4 and Equation 9 for a soil with f_{oc} = 0.0125 (OM = 2%) Kd's of 1, 20, 40, and 1000 would represent log K_{ow}'s of 2.3, 3.6, 3.9, and 5.3, respectively.

Liquid-Vapor Partition

Vapor phase partitioning of a compound in the soil influences the spread of the compound through the soil. Even for chemicals with relatively low vapor pressure, this transport route has been shown to be significant (Mayer,et al., 1974). Those chemicals that have a high vapor pressure may easily move from the soil solution into the soil air phase, where they can move throughout the soil

and across the soil surface. The vapor-phase may be taken up by the plant either through roots or by above ground portions of the plant.

The compartmentalization of the compound between the soil solution and the air spaces in the soil is frequently described by Henry's Law (Jury et al., 1983) with the extent of partitioning described by Henry's Constant (Hc). This can be calculated as:

$$\text{Henry's Constant (Hc)} = \frac{16.04 P M}{T S} \qquad [11]$$

where P = vapor pressure of pure solute in mm/Hg,
M = molecular weight of solute,
T = absolute temperature, and
S = solubility in water mg/L

(Thibodeaux, 1979). Henry's Constant may be expressed in different units and vary by several orders of magnitude depending upon the source of the original data. For example, estimated values for vinyl chloride of 2.3×10^{-2} to 6.39 atm m^3/mol are reported by Mackay and Shiu (1981) and Goldstein (1982), respectively. Experimentally determined Hc values are considered more reliable than calculated values. Henry's Constant, dimensionless, for the priority pollutants is provided in Table 1.

Comprehensive studies have not been conducted to determine the Hc above which volatilization plays an important role in the transport of a chemical in the atmosphere. Thus, it is not possible to select a Hc above which transport in the soil will occur primarily in the vapor phase. However, a partition between the vapor and aqueous phases of greater than 10^{-4} is normally sufficient for a chemical to be a good preemergence herbicide (Graham-Bryce, 1984). Jury et al.,(1984) utilized three volatility categories with Hc values of 2.5×10^{-3}, 2.5×10^{-5} and 2.5×10^{-7}. Gillett (1983) utilized values of 10^{-3} and 6×10^{-5} in his classification. Thus, the value of 10^{-4} may be a reasonable transition point for determining when vapor diffusion becomes important. This would mean that vapor diffusion would be important for all PCB's and halogenated aliphatics and unimportant for some of the monocyclic and polycyclic aromatics and many pesticides. Soil sorption can significantly reduce chemical volatilization (Fairbanks et al., 1987) thus, the arbitrary value of 10^{-4} may overestimate the importance of volatilization in high organic carbon soils. Jury et al., (1983) used Hc and K_{oc} to calculate volatilization flux from soil.

PLANT UPTAKE OF ORGANIC CHEMICALS

Chemical uptake by plants is a complex process that may involve a compound specific active processes, and/or a passive process in which the chemical accompanies the transpiration water through the plant. If the former case dominates, a rigorous relationship between plant uptake and the chemicals

physicochemical parameters may not exist, although some general guidelines may be expected. If uptake into the plant is a passive process, rigorous relationships should exist.

It is generally accepted that there are four main pathways by which a chemical in the soil can enter a plant (Topp et al., 1986). These are:

1. root uptake and subsequent translocation by the transpiration stream,
2. vegetative uptake of vapor from the surrounding air,
3. uptake by external contamination of shoots by soil and dust, followed by retention in the cuticle or penetration through it, and
4. uptake and transport in oil cells which are found in oil containing plants like carrots and cress.

The amount of an organic chemical found in a plant will be the sum total of each of these transport routes minus metabolic losses. Their respective importance will depend upon the nature of the organic chemical, the nature of the soil, and the environmental conditions under which plant exposure occurs. Pathways 3 & 4 are significant only in specific situations. Thus, for the purpose of describing the general case of plant uptake, they can be discounted as major routes of plant contamination. Most reported instances of plant uptake of soil-borne organic compounds make no attempt to discriminate between pathways 1 & 2. Therefore, the relative importance of each pathway, under different environmental conditions, has not been assessed at present.

<u>Root Uptake And Translocation</u>

Shone and Wood (1972) investigated the absorption and translocation of the herbicide simazine by 6-day-old barley plants in solution cultures. The experiments were either 24- or 48-hour experiments conducted under different conditions of humidity, light intensity, temperature, and levels of metabolic inhibitors. The relationship between simazine transport and water uptake was described by a transpiration stream concentration factor (TSCF), defined as:

$$\text{TSCF} = \frac{\mu\text{g simazine in shoots per mL water transpired}}{\mu\text{g simazine per mL of external solution}}$$

They found that water was taken up preferentially to simazine, because the TSCF was always less than unity, i.e., the concentration of simazine in the plant shoots per mL of water transpired never reached that in the external solution. There was no evidence of loss of or breakdown of the parent compound during the experiment. The concentration of simazine in the plant roots, on a fresh weight basis, however, reached a value greater than unity as a result of physical sorption of the herbicide to the root tissue.

Evaluation of other triazines led to the conclusion that plant uptake was, in general, a passive process because TSCF was less than unity, (Shone et al.,1973).

Plant uptake of 6 herbicides and a fungicide showed that TSCF was independent of concentration and less than unity for all except 2,4-D at pH 4.0 (Shone and Wood, 1974). In the case of 2,4-D at pH 4.0, plant uptake was metabolically influenced. Briggs et al.,(1982) evaluated plant uptake of 18 chemicals and found that the TSCF was less than unity for all chemicals studied. They related the TSCF to the octanol/water partition coefficient (K_{ow}) for the chemicals and found a bell shaped relationship between TSCF and K_{ow}, with a broad maximum around a K_{ow} of 1.8. A Gaussin curve (Figure 5) was fitted to the data such that:

$$TSCF = 0.784e^{-[(\log K_{ow} - 1.78)^2/2.44]} \qquad [12]$$

The authors suggested that at K_{ow} values below 1.8, translocation is limited by the lipid membranes in the root. At K_{ow} values above 1.8, translocation is limited by the rate of transport of the lipophilic chemical from the plant root to the top of the plant. All the TSCF values were below unity, suggesting passive chemical movement into the shoot with the transpiration stream. There was no evidence that chemicals were taken up against a concentration gradient.

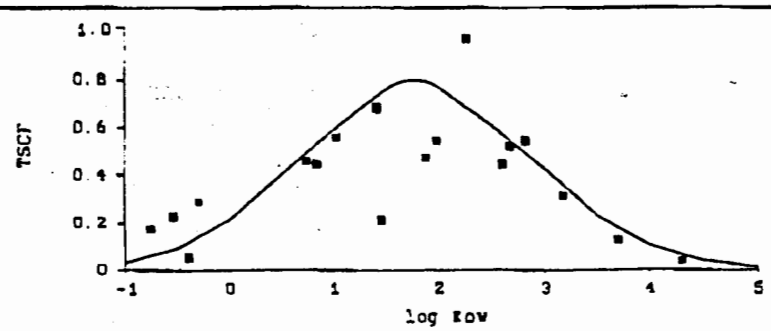

FIGURE 5 RELATIONSHIP BETWEEN log Kow AND TRANSPIRATION STREAM CONCENTRATION FACTOR
Adapted from Briggs et al. 1982

Shone and Wood (1974) proposed that the uptake of a chemical into a plant root could be described by a root concentration factor (RCF), defined as:

$$RCF = \frac{\text{concentration in root, } (\mu g/g \text{ fresh wt.})}{\text{concentration in external solution, } (\mu g/mL)}$$

Using radiolabelled herbicides in solution culture with barley seedlings, they showed that the quantity of the herbicide transported to the plant stems (TSCF) could not be inferred from the concentration in the plant roots (RCF). In addition, although the RCF of some of the tested herbicides exceeded unity, uptake was not affected by temperature. This, suggests the compounds were retained by physical sorption rather than biochemically.

When barley seedlings were transferred from the herbicide amended solution culture to a herbicide free solution, RCF decreased before TSCF was affected by

the change (Shone et al., 1974). Thus lipophilic herbicides appear to penetrate the cortical cells of the root whereas the lipophobic herbicides are largely confined to the free cell space in the root.

Briggs et al.,(1982) found that RCF was related to K_{ow}. Starting with a value of less than unity for polar compounds, RCF increased with increasing K_{ow}. Sorption of chemicals by macerated roots was very closely related to the RCF of living roots, for the more lipophilic chemicals. In contrast, the RCF of macerated roots continued to decrease as the lipophilicity decreased (Figure 6). There was a linear relationship between the log concentration factor of the macerated roots and log K_{ow}:

$$\log RCF_{(\text{macerated root})} = 0.77 \log K_{ow} - 1.52 \qquad [13]$$

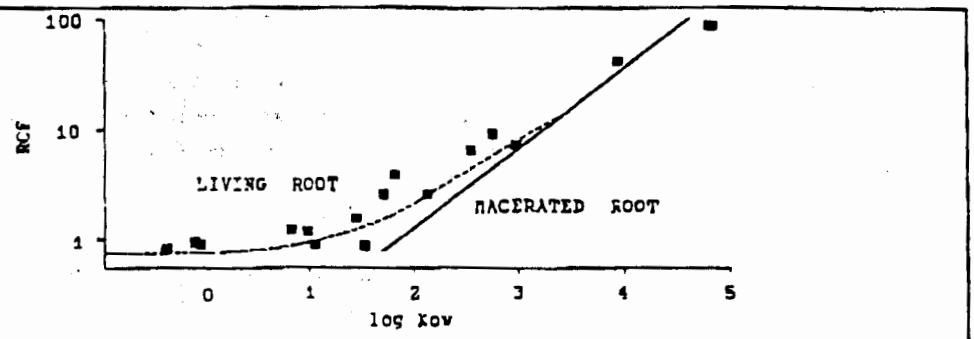

FIGURE 6 EFFECT OF TISSUE STATUS ON THE RELATIONSHIP BETWEEN log Kow AND ROOT CONCENTRATION FACTOR
Adapted from Briggs et al. 1982

Assuming that RCF of living roots could be explained by two processes: (1) a partitioning of the organic chemical between the lipophilic root tissue and external solution culture and (2) a fraction of root that is aqueous and equal in concentration to external solution phase (constant for all compounds, 0.82). Briggs et al., (1983) suggested that sorption of chemicals by the root is a partitioning described by:

$$\log(RCF - 0.82) = 0.77 \log K_{ow} - 1.52 \qquad [14]$$

∴ log RCF = 0.77 log Kow − 1.52 + 0.82

They proposed an analogous stem concentration factor (SCF):

$$SCF = \frac{\text{concentration in stem } (\mu g/g \text{ fresh wt.})}{\text{concentration in external solution } (\mu g/mL)}$$

Macerated stems sorption of organic compounds was also related to the K_{ow} of the compound:

$$\log SCF_{(\text{macerated stem})} = 0.95 \log K_{ow} - 2.05 \qquad [15]$$

Assuming that the contribution of the aqueous phase in the stem was similar to that in roots (0.82), the partition between the stem and xylem stream is:

$$\log(K_{(\text{stem/xylem sap})} - 0.82) = 0.95 \log K_{ow} - 2.05 \qquad [16]$$

The SCF is then given by the $K_{(stem/xylem\ sap)}$ partition coefficient multiplied by the partition of the external solution present in the xylem sap (TSCF):

$$SCF = [10^{(0.95\log K_{ow} - 2.05)} + 0.82] * [(0.784)10^{-0.434[(\log K_{ow} - 1.78)^2/2.44]}] \quad [17]$$

For 15 chemicals ($\log K_{ow}$ from -0.57 to 3.7), the experimental points fit the predicted line quite well (Figure 7). The shift in log K_{ow} where TSCF reaches a maximum (1.8) to where SCF reaches a maximum (4.5) arises because sorption of the more lipophilic compounds by the stem tissue increases faster than the TSCF decreases. The predicted decline in SCF for compounds of log K_{ow} > 4.5 was not tested.

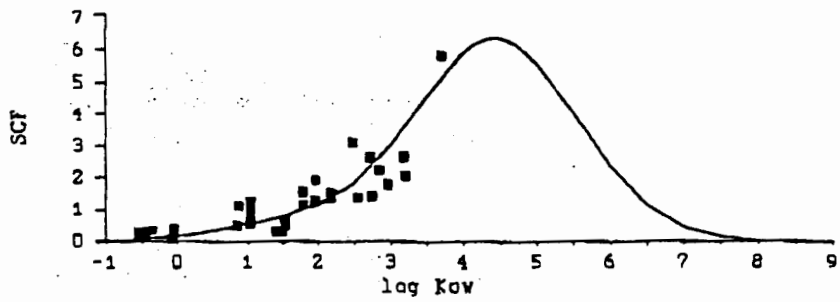

FIGURE 7 RELATIONSHIP BETWEEN log Kow AND PLANT CONCENTRATION FACTOR

adapted from Briggs et al. 1983

There have been other attempts to relate plant uptake and translocation of an organic chemical to either the physical or chemical properties of the chemical. Topp et al., (1986) reported the relationship:

$$\log RCF = 0.63 \log K_{ow} - 0.959 \quad [18]$$

following their exposure of barley seedlings for 7-days to various chemicals in water culture.

The concentration factor (CF) concept is a useful way of describing the relative concentration of an organic chemical in a particular plant part. It has many limitations, however. These arise because the concentration of organic chemicals, both within the soil or nutrient solution and within the plant part do not remain constant with time. Chemicals in the soil, or in nutrient solution, may be depleted by plant uptake or degradation; chemicals in a plant may also be reduced with time by degradation within the plant, or by increases in plant mass effectively diluting the chemical. Changes in uptake as measured by the CF, have been reported, Figure 8 (Topp et al., 1986). Different CF's arise depending upon the timing of the actual sampling. Further it seems logical that the CF would depend upon soil concentration, initial vs soil concentration at time of plant

sampling. Further research on this topic is needed to define the effect of time of sampling (both plant and soil) on CF's so different experiments can be compared.

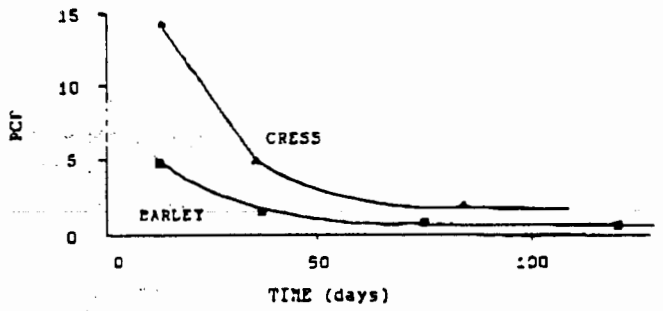

FIGURE 8 EFFECT OF PLANT TYPE AND LENGTH OF GROWTH PERIOD ON THE PLANT CONCENTRATION FACTOR
Adapted from Topp et al. 1986

The work of Shone, Briggs, and their co-workers reported above was carried out in nutrient solution cultures where sorption and desorption effects of soil organic matter were absent. The application of their results to plant uptake from field soils requires that soil sorption be considered. The effect of soil sorption on soil solution concentration can be mathematically described using the following relationship:

$$\delta C_T = \delta C_s + \theta C_L \qquad [19]$$

where C_T is the total organic chemical concentration in the soil (μg/g), δ is the soil bulk density (g/cm^3), C_s is the adsorbed chemical concentration (μg/g), θ is the soil-water content by volume (mL/cm^3), and C_L is the chemical concentration in the soil-water phase (μg/mL). Using the linear equilibrium relationship in Equation 3 and 4 allows Equation 19 to be rewritten in terms of C_L such that:

$$\frac{C_L}{C_T} = \frac{\delta}{\delta K_{oc} f_{oc} + \theta} \qquad [20]$$

It is now possible to combine equations relating soil sorption and soil solution concentration and calculate RCF, TSCF, and SCF for different chemicals on a total soil concentration basis. Substituting Equation 20 into Equation 17 where C_L is the external solution and:

$$SCF_{(SOIL)} = \frac{\text{concentration in stem}}{\text{concentration in soil}}$$

gives:

$$SCF_{(SOIL)} = \frac{\delta}{\delta K_{oc} f_{oc} + \theta} \left([10^{(0.95 \log K_{ow} - 2.05)} + 0.82] - [(0.784) 10^{-0.434[(\log K_{ow} - 1.78)^2 / 2.44]}] \right) \qquad [21]$$

For nutrient solutions this equation reduces to Eq [17] when $f_{oc} = 0$, $\theta = 1$, and $\delta = 1$. Inclusion of soil sorption into the SCF from Briggs et al.,(1983)

alters the relationship between SCF and log K_{ow} such that the log K_{ow} where plant adsorption is a maximum decreases from 4.5 for nutrient solution to 1 for soils. (Figure 9). The decrease in SCF for chemicals with log K_{ow} greater than 1 is supported by the published literature on plant uptake in soil systems (Travis and Arms, 1988).

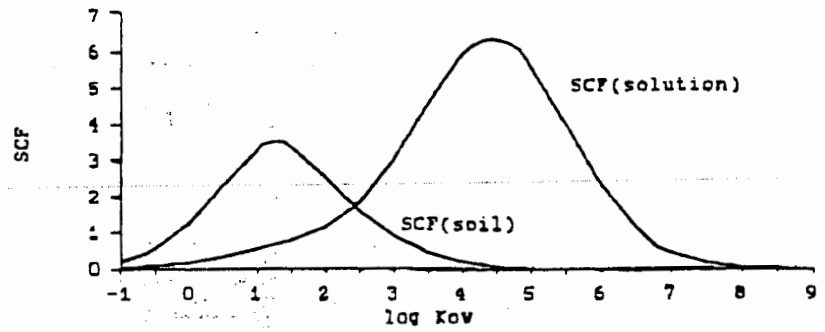

FIGURE 9 EFFECT OF SOIL ON THE RELATIONSHIP BETWEEN log Kow AND STEM CONCENTRATION FACTOR

Equation 21 implies that plant uptake is related to soil organic matter content (Figure 10). Differences in the plant uptake of an organic chemical in soils with different organic carbon contents has been shown experimentally. Lichtenstein et al., (1967) for example, showed higher concentrations of the pesticide aldrin in roots of peas when grown in aldrin- polluted quartz sand compared to a loam soil containing approximately the same total concentration of the pollutant.

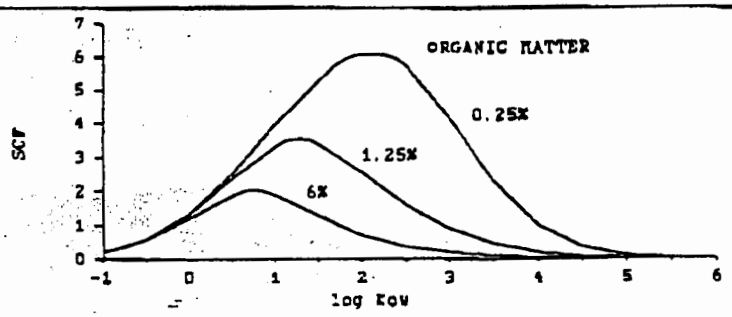

FIGURE 10 EFFECT OF SOIL ORGANIC MATTER ON THE RELATIONSHIP BETWEEN log Kow AND STEM CONCENTRATION FACTOR

It is also apparent from Equation 21 that increases in soil water content reduce SCF (Figure 11). However, for a soil with a f_{oc} of 0.0075 (1.25% organic matter), changes in soil water content over the range 0.1 to 0.5 mL/cm^3 altered SCF less than 10% for chemicals with a K_{ow} greater than 2.5. The fraction in solution, ($\theta C_l/C_T$), increases as soil water content increases even though the organic chemical concentration (C_l) in the soil solution-phase decreases.

Therefore, if plant transpiration were increased by increasing soil water content, plant concentration could be increased. Walker, (1971) found that the phytotoxicities of the pesticides atrazine, simazine, linuron, lenacil, and aziprotryne were increased as the moisture content of the soil increased. He related the effect to differences in the quantities of the pesticides that were accumulated by the plants, with the degree of accumulation being directly proportional to water uptake.

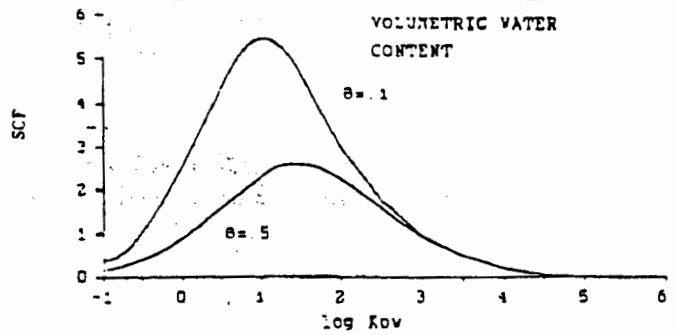

FIGURE 11 EFFECT OF SOIL WATER CONTENT ON THE RELATIONSHIP BETWEEN log Kow AND STEM CONCENTRATION FACTOR

In conclusion, assuming degradation of the organic chemical does not occur within the plant, and plant root uptake and translocation of organic chemicals from the soil is passive, plant uptake can be described as a series of consecutive partitions reactions. Partitioning occurs between soil solids and soil water, soil water and plant roots, plant roots and transpiration stream, and transpiration stream and plant stem. This partitioning can be related to the K_{ow} of organic compounds such that pollutants with high log K_{ow} values, (eg. TCDD (6.14), PCB's (4.12-6.11), some of the phthalate esters (above 5.2) and the polycyclic aromatic hydrocarbons (4.07-7.66)) are most likely to be sorbed by the soil and/or plant root. Chemicals with lower K_{ow} values are likely to be translocated within the plant and may reach significant concentrations within the above ground portions of the plant.

Vapor Phase Uptake

For volatile compounds, diffusion in the vapor phase and subsequent uptake by the root and/or shoot may be an important route of chemical entry into plants (Parker, 1966, and Prendeville, 1968). Two processes precede the penetration of chemicals in the soil into plant tissue via the air: 1) volatilization of the chemical from the soil and 2) deposition from the air onto the plant surface. Soil volatilization depends upon the vapor pressure of the compound which varies according to ambient temperatures, water solubility of the compound, and sorption capacity and physical properties of the soil.

Increasing the soil-water content of a soil will increase the potential for volatilization loss of a chemical (Guenzi and Beard, 1970). Harris and Lichtenstein (1961) showed that the rate of volatilization of aldrin from soil increased with aldrin concentration, soil moisture, relative humidity, temperature and the rate of air movement. Chemical concentration effects cease when the concentration reaches that required to give a maximum saturation vapor density equivalent to that of the pure compound. For dieldrin in a Gila silt loam soil this concentration was 25 ppm (Farmer et al., 1972). These authors also report that under similar environmental conditions the rate of volatilization was lindane > dieldrin > DDT, which is the same order for increasing vapor pressures. Jury et al., (1983 and 1984) developed a behavior assessment model that separates compounds into volatilization categories based on Henry's constants.

There have been few investigations aimed at separating root uptake and translocation of a chemical from vapor phase uptake into plant shoots. In an experiment designed to discriminate these effects, Beall and Nash (1971) found soybean shoots were contaminated by soil applied dieldrin, endrin and heptachlor largely via root uptake and subsequent translocation. Vapor phase foliar sorption however dominated for DDT and was nearly 7 times greater than root sorption and translocation. Foliar contamination from vapor sorption of residues from all four insecticides was similar (about 6.5 ppm plant dry weight), whereas contamination from root sorption and translocation varied from 38 ppm to 1 ppm depending upon the compound.

Using similar experimental techniques, Fries and Marrow (1981) found that PCBs reached the shoots of plants via the vapor phase rather than from root uptake, although the importance of this route for PCB contamination of plants remains inconclusive.

Topp et al., (1986) investigated the uptake of 16 organic chemicals by barley seedlings. Foliar uptake was related to the amount of chemical volatilized from the soil surface. The relationship (Figure 12) after 7 days exposure was:

$$FU = 46.11 + 28.95 \log VOL \qquad [22]$$

where FU was foliar uptake as percent of total ^{14}C uptake, and VOL was the organic ^{14}C trapped from the air plus that sublimated on the walls of the exposure chamber as percent of the total ^{14}C applied (Note that in the original publication the sign in front of log VOL is negative, this is assumed to be a typographical error). Four compounds (benzene, pentachlorophenol, diethylhexylphthalate, and the phenylenediamine pigment) did not fit the calculated line because they were nonpersistent and taken up after mineralization to $^{14}CO_2$.

There are many difficulties in extrapolating vapor phase uptake in the

laboratory to that in the field. Overall, volatilization rates are likely to be higher in the laboratory than in the field. This is because laboratory soils are normally kept moist to encourage plant growth, and this encourages volatilization. In addition, the actual deposition of volatilized chemicals onto a plant in the field is likely to be lower as atmospheric turbulence may be higher.

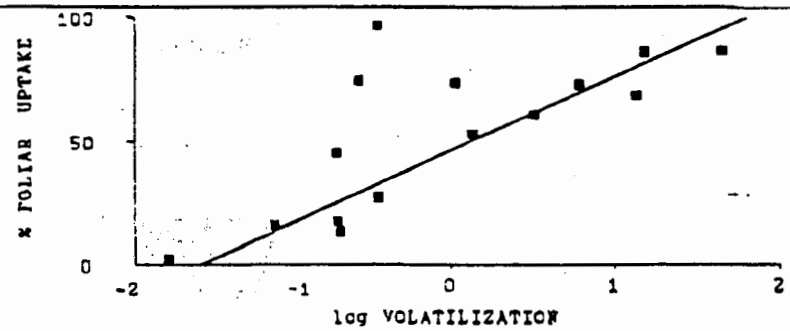

FIGURE 12 RELATIONSHIP BETWEEN VOLATILIZATION AND FOLIAR UPTAKE Adapted from Topp et al. 1986

The importance of plant uptake of organic chemicals via volatilization under field conditions, remains to be determined. However, it appears potentially significant for compounds with Hc greater than 10^{-4}. The impact could be estimated by calculating cumulative volatilization losses over the growing period and assuming that all of it ends up in the plant. The model of Jury et al., 1983 could be utilized for this purpose.

Plant Type

The final variable affecting plant uptake of soil-borne organic pollutants is the plant species itself. There has been no systematic examination of plant responses to organic chemicals in soil, although it does appear that, as with plant uptake of soil-borne heavy metals, there is variation in uptake both between species and within the same species on an individual level (Chaney, 1985; and McNeilly, 1978). For example: Harris and Sans (1967) found that sugar beat roots accumulated more dieldrin from a clay soil that contained dieldrin, than did carrots, potatoes, sugar beet tops, corn, oats, and alfalfa. Lichtenstein and Shulz (1965) on the other hand, report that carrots usually take up more organochlorine insecticides than do other root crops such as potatoes, radish, turnip, and beet. This apparent contradiction can be resolved by consideration of varietal differences which can be as much as 400% when different carrot varieties are grown in soil containing endrin (Hermanson et al., 1970).

CONCLUSIONS

In solution culture, the movement of nonionic organic compounds into roots is a passive process, equivalent to a partitioning between the liquid and solid

phase, and can be related to the octanol water partition coefficient of the compound. Subsequent translocation of the chemical from roots to shoots depends on the K_{ow} of the compound and the transpiration rate of the plant. Based on available data, compounds with a log K_{ow} of approximately 4.5 are most likely to accumulate in the stem and leaf tissue of plants.

TABLE
plant

In soil systems, there is competition between the plant and soil solids (organic fraction) for the partitioning of organics from solution. As the sorption of the compound by the soil organic phase increases, the quantity available for plant uptake decreases. Based upon these considerations compounds with log K_{ow} of 1 -2 are most likely to have significant transport of the chemical to above ground plant tissue produced in soil systems. If metabolism of the compound in the roots is significant, even compounds with low log K_{ow}'s may not be translocated (McFarlen et al., 1987). Compounds with high log K_{ow} > 5.0 would not be expected to be present in above ground plant tissue if plant uptake is limited by soil solution.

The potential for root or plant sorption of organic compounds from vapor is dependent upon the vapor pressure of the compound. Very few experiments on this route of plant contamination have been conducted. Based upon the movement of herbicides in the soil, a Henry's constant of 10^{-4} may be used as a transition point between primary movement in solution and vapor phases. If it can be assumed that vapor movement in the soil will result in vapor uptake by the plant, then those compounds with Hc $>10^{-4}$ are potential candidates for vapor phase uptake.

Superimposed upon both of these processes is the half-life of the compound. If it is short, i.e., less than 10 days, the chemical is likely lost from the system before it can be taken up by the plant. Those compounds with long half lives, i.e., greater than 6 months or greater than the growing season of the plant, presist long enough to impact plants.

Applying these screening processes to the priority pollutants, listed in Table 1, reduces the number of chemicals likely taken up by plants. For example, if plant uptake and translocation without vaporization is the pathway of concern, the list of 107 chemicals is reduced to 50 on the basis of half-life and K_{ow}, (Table 2). If vaporization is of concern the list is reduced from 107 to 64 on the basis of half-life and Hc, (Table 3).

TABLE 2 Log K_{ow}, Half-life and Hc for Priority Pollutants which are subject to plant uptake from soil

Compound	log K_{ow}	$T_{1/2}$	Hc	Compound	log K_{ow}	$T_{1/2}$	Hc
PESTICIDES							
20.Acrolein	-0.09	B	2.8E+03	26.Dieldrin	2.9	C	3.0E+06
27.Endosulfan	3.55	C	nd	30.Heptachlor epoxide	3.9	C	3.2E+05
31.Hexachlorocyclohexane	3.8	B	3.0E-04	32.Lindane	3.72	C	6.0E-04
33.Isophorone	1.70	nd	nd	35.Toxaphene	3.85	C	2.1E+01
POLYCHLORINATED BIPHENYLS							
HALOGENATED ALIPHATIC HYDROCARBONS							
38.Chloromethane	0.91	C	1.6E+01	39.Dichloromethane	1.25	B	8.5E-02
40.Trichloromethane	1.9	B	1.2E+01	41.Tetrachloromethane	2.64	nd	9.6E+01
42.Chloroethane	1.54	B	6.1E+01	43.1,1-dichloroethane	1.79	B	1.7E+01
44.1,2-dichloroethane	1.48	B	3.8E-02	45.1,1,1-trichloroethane	2.17	nd	1.3E+00
46.1,1,2-trichloroethane	2.17	nd	3.1E+01	54.1,2-dichloropropane	2.28	nd	1.2E+01
56.Hexachlorobutadiene	3.74	C	4.3E-01	58.Bromomethane	1.10	B	4.4E+00
59.Bromodichloromethane	1.88	nd	nd	60.Dibromochloromethane	2.09	nd	nd
61.Tribromomethane	2.30	nd	2.4E-02	62.Dichlorodifluoromethane	2.16	C	6.3E+01
63.Trichlorofluoromethane	2.53	nd	2.4E+00				
HALOGENATED ETHERS							
65.Bis(2-chloroethyl)ether	1.58	nd	4.7E-05	66.Bis(2-chloroisopropyl)ether	2.58	nd	4.7E-02
70.Bis(2-chloroethoxy)methane	1.26	C	1.1E-05				
MONOCYCLIC AROMATICS							
72.Chlorobenzene	2.84	nd	1.5E-01	73.1,2-dichlorobenzene	3.38	nd	1.5E-01
74.1,3-dichlorobenzene	3.55	nd	1.1E-01	75.1,4-dichlorobenzene	3.55	nd	9.9E-02
79.Nitrobenzene	1.85	C	5.4E-04	81.2,4-dinitrotoluene	2.01	nd	1.3E-02
82.2,6-dinitrotoluene	2.05	nd	1.3E-02	84.2-chlorophenol	2.17	nd	1.9E-04
86.2,4,6-trichlorophenol	3.61	nd	1.7E-04	88.2-nitrophenol	1.76	C	3.2E-04
89.4-nitrophenol	1.91	B	2.6E-04	90.2,4-dinitrophenol	1.53	C	2.7E-08
91.2,4-dimethylphenol	2.50	nd	7.5E-04	92.p-chloro-m-cresol	3.11	nd	1.0E-04
93.4,6-dinitro-o-cresol	2.85	nd	nd				
PHTHALATE ESTERS							
94a.Dimethyl phthalate esters	2.12	B	8.8E-06	94b.Diethyl	3.22	B	1.9E-03
POLYCYCLIC AROMATIC HYDROCARBONS							
95d.Naphthalene	3.37	C	2.0E-02				
MISCELLANEOUS COMPOUNDS							
99.Dimethyl nitrosamine	0.06	nd	nd	100.Diphenyl nitrosamine	2.37	nd	nd
101.Di-n-propyl nitrosamine	1.31	nd	nd	104.1,2-diphenylhydrazine	3.03	nd	nd

TABLE 3. Log K_{ow}, Half-life and Hc for the Priority Pollutants which are subject to plant uptake via volatilization

Compound	log K_{ow}	$T_{1/2}$	Hc	Compound	log K_{ow}	$T_{1/2}$	Hc
PESTICIDES							
20.Acrolein	-0.09	B	2.8E-03	22.Chlordane	4.3	C	3.9E-03
23.DDD	5.99	C	0.5E+00	25.DDT	5.98	C	2.0E-03
27.Endosulfan	3.55	C	nd	31.Hexachlorocyclohexane	3.8	B	3.0E-06
33.Isophorone	1.70	nd	nd	34.TCDD	6.14	C	nd
35.Toxaphene	3.85	C	2.1E-01				
POLYCHLORINATED BIPHENYLS							
36a.Aroclor 1016	4.38	C	8.6E+01	36b.Aroclor 1221	4.09	C	1.3E-02
36c.Aroclor 1232	4.54	C	2.1E+00	36d.Aroclor 1242	4.11	C	5.5E-02
36e.Aroclor 1248	5.6	C	1.1E-01	36f.Aroclor 1254	6.04	C	1.1E-01
36g.Aroclor 1260	6.11	C	2.9E-01	37.2-chloronaphthalene	4.12	C	1.3E-02
HALOGENATED ALIPHATIC HYDROCARBONS							
38.Chloromethane	0.91	C	1.6E+01	39.Dichloromethane	1.25	B	8.5E-02
40.Trichloromethane	1.9	B	1.2E-01	41.Tetrachloromethane	2.64	nd	9.6E-01
42.Chloroethane	1.54	B	6.1E-01	43.1,1-dichloroethane	1.79	B	1.7E-01
44.1,2-dichloroethane	1.48	B	3.8E-02	45.1,1,1-trichloroethane	2.17	nd	1.3E+00
46.1,1,2-trichloroethane	2.17	nd	3.1E-01	48.Hexachloroethane	4.62	nd	4.1E-01
54.1,2-dichloropropane	2.28	nd	1.2E-01	56.Hexachlorobutadiene	3.74	C	4.3E-01
58.Bromomethane	1.10	B	4.4E+00	59.Bromodichloromethane	1.88	nd	nd
60.Dibromochloromethane	2.09	nd	nd	61.Tribromomethane	2.30	nd	2.4E-02
62.Dichlorodifluoromethane	2.16	C	6.3E+01	63.Trichlorofluoromethane	2.53	nd	2.4E+00
HALOGENATED ETHERS							
66.Bis(2-chloroisopropyl)ether	2.58	nd	4.7E-02	68.4-chlorophenyl phenylether	4.08	nd	1.0E-02
69.4-bromophenyl phenyl ether	4.28	nd	nd				
MONOCYCLIC AROMATICS							
72.Chlorobenzene	2.84	nd	1.5E-01	73.1,2-dichlorobenzene	3.38	nd	1.5E-01
74.1,3-dichlorobenzene	3.53	nd	1.1E-01	75.1,4-dichlorobenzene	3.53	nd	9.9E-02
77.Hexachlorobenzene	6.18	C	7.0E-02	81.2,4-dinitrotoluene	2.01	nd	1.3E-02
82.2,6-dinitrotoluene	2.05	nd	1.3E-02	93.4,6-dinitro-o-cresol	2.85	nd	nd
PHTHALATE ESTERS							
94b.Diethyl	3.22	B	1.9E-03				
POLYCYCLIC AROMATIC HYDROCARBONS							
95a.Acenaphthene	4.13	C	1.0E-02	95b.Acenaphthylene	4.07	C	4.8E-03
95c.Fluorene	4.18	C	4.8E-03	95d.Naphthalene	3.37	C	2.0E-02
96a.Anthracene	4.45	C	1.1E-02	96c.Phenanthrene	4.46	C	1.6E-03
97b.Benzo(b)fluoranthene	6.57	nd	nd	97c.Benzo(k)fluoranthene	6.84	C	nd
97d.Chrysene	5.61	C	8.8E-02	97e.Pyrene	5.32	C	2.8E-01
98a.Benzo(ghi)perylene	7.23	C	nd	98b.Benzo(a)pyrene	6.04	C	4.9E-01
98c.Dibenzo(a)anthracene	5.97	C	nd	98d.Indeno(123-cd)pyrene	7.66	C	nd
MISCELLANEOUS COMPOUNDS							
99.Dimethyl nitrosamine	0.06	nd	nd	100.Diphenyl nitrosamine	2.57	nd	nd
101.Di-n-propyl nitrosamine	1.31	nd	nd	104.1,2-diphenylhydrazine	3.03	nd	nd

Clearly, plant uptake of soil borne organic pollutants is a complex phenomena. More work is needed before the potential environmental impact of organic pollutants can be adequately assessed and actions designed to limit such impacts.

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