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ARTICLE *in* ENVIRONMENTAL SCIENCE AND TECHNOLOGY · DECEMBER 1977

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velocity and wind direction at low to moderate wind speeds are not sufficiently accurate for modeling purposes. The ground temperature, terrain, and low level objects tend to introduce very large fluctuations in these variables. Analysis shows that the signal-to-noise ratio is small—not a desirable characteristic for modeling purposes.

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Received for review August 11, 1975. Accepted November 29, 1976.

Partition Coefficient and Bioaccumulation of Selected Organic Chemicals

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■ An empirical equation is established to relate the experimental *n*-octanol/water partition coefficients to the aqueous solubilities of a wide variety of chemicals including aliphatic and aromatic hydrocarbons, aromatic acids, organochlorine and organophosphate pesticides, and polychlorinated biphenyls. Such a correlation, covering more than eight orders of magnitude in solubility (from 10⁻³ to 10⁴ ppm) and six orders of magnitude in partition coefficient (from 10 to 10⁷), allows an assessment of partition coefficient from solubility with a predicted error of less than one order of magnitude. A correlation is observed between the bioconcentration factors in rainbow trout and the aqueous solubilities for some stable organic compounds.

The current emphasis on the environmental impact of both agricultural and industrial chemicals has resulted in an increased reliance on the physical and chemical properties of these compounds to assess their environmental behavior. The *n*-octanol/water partition coefficient, for example, has proved useful as a means to predict soil adsorption (6), biological uptake (18), lipophilic storage (10), and biomagnification (21, 24, 25, 27). Unfortunately, the partition coefficients of many compounds of environmental significance are not always available despite a recent extensive compilation (20). Assessment of partition coefficients from a more readily available physical parameter would be useful.

By definition, the partition coefficient expresses the equilibrium concentration ratio of an organic chemical partitioned between an organic liquid (e.g., *n*-octanol) and water. This partitioning is, in essence, equivalent to partitioning an organic chemical between itself and water. Consequently, one would suspect that a correlation might exist between the partition coefficient and the aqueous solubility. An equivalent correlation has been demonstrated earlier by Hansch et al. (13) for several classes of organic liquids of relatively low molecular weights whose aqueous solubilities generally fall in the high ppm (mg/L) range. In the report of Hansch et al., however, most partition coefficients of the organic liquids were assessed from the π constants of the substituents in relation to some parent compounds since the experimental values were not determined.

It would be of interest to test this correlation using the experimental data on various types of pesticides and industrial chemicals. This correlation would be useful as it serves not only as an indication of the interrelationship between these

two physical parameters but also as a convenient means to predict a reasonable partition coefficient from the aqueous solubility. For example, it has been noted that partition coefficients reported for a given pesticide (e.g., DDT) often differ more than several orders of magnitude (21, 28, 33). The use of this correlation will help clarify questionable data. In our present studies we are reporting a satisfactory relationship between measured partition coefficients and aqueous solubilities for a wide variety of chemicals. By combining the relationships of partition coefficient to aqueous solubility and to biomagnification (21, 24, 25, 27), a correlation between biomagnification and aqueous solubility as reported by Lu and Metcalf (21) and Metcalf et al. (25) is supported. In the present report, a similar correlation is demonstrated for several stable organic compounds using the biomagnification data in rainbow trout from Neely et al. (27).

Experimental

The aqueous solubilities of 2,4,5,2',5'-PCB, 2,4,5,2',4',5'-PCB, and six organophosphate pesticides were determined at room temperature. The method consisted of equilibrating a quantity of the compound with water by determining the concentration at different time intervals until saturation was achieved. The water used in the solubility study was passed through a 1 × 7 cm XAD-2 column to remove organic impurities. The chemical was dissolved in approximately 10 mL distilled ether and swirled onto the walls of a flask, evaporating the ether and leaving the compound deposited on the walls. Water was added and stirred slowly to avoid disturbing the coated walls. Aliquots of the aqueous solution were removed via a sintered-glass filter immersed in the solution (16) and analyzed at regular intervals with GLC until no further change in concentration was noticed. Preliminary values of the water solubilities of 4,4'-PCB and *p,p'*-DDE were also determined under similar conditions except that samples taken for analysis were equilibrated for a shorter time interval. The determined solubilities for the latter two compounds, however, are in reasonably good agreement with the reported literature values (3, 34).

The partition coefficients of the three PCB's and 12 organophosphates were determined by the following method. A stock solution of approximately 1 mg/mL compound in *n*-octanol was prepared. A 2-mL aliquot and 20 mL of water were shaken for 16 h at 20 °C in a closed 25-mL Corex centrifuge tube. After equilibration and centrifugation (17 500 rpm, 37 000 G at 20 °C for 20 min), the two phases were separated and extracted or diluted as necessary to analyze by GLC. The

n-octanol used was distilled several times to remove trace amounts of impurities which tended to produce emulsions. Formation of emulsions gives poor phase separation and results in a wide scatter in the partition coefficients. It is essential to analyze the concentrations in both phases for a compound of low aqueous solubility when determining the partition coefficient, since the compound may adsorb on the glass walls of the container. All compounds were analyzed on a Tracor 550 GLC equipped with a Ni⁶³ electron capture detector. The columns used were as follows: DDT and PCB's—a mixed phase 1.5% OV-17/1.95% OV-210 on 60/80 Gas-Chrom Q, 2 mm × 2 ft glass column; organophosphates—5% OV-225 on 60/80 Gas-Chrom Q, 2 mm × 2 ft glass column or >0.2% Carbowax 20M on 80/100 Chromasorb W, 2 mm × 2 ft glass column (1).

Results and Discussion

Table I shows a list of the experimental values for the aqueous solubilities and *n*-octanol/water partition coefficients for various types of chemicals. A plot of log (partition coefficient) vs. log (solubility in water) using the experimental values obtained in this laboratory as well as values cited in the literature is shown in Figure 1. The solubility values from the literature were taken from a temperature range of 10–30 °C and the partition coefficient values at room temperature. Our

solubility and partition coefficient values were determined at 20 °C except the solubilities of the two PCB's which were done at 24 °C. A satisfactory linear relationship is observed between the log (partition coefficient) and log (aqueous solubility), which extends to more than eight orders of magnitude in solubility (10⁻³ to 10⁴ ppm) and six orders of magnitude in partition coefficient (10 to 10⁷). The regression equation shows:

$$\log K = 5.00 - 0.670 \log S \quad (1)$$

where *K* = *n*-octanol/water partition coefficient and *S* = aqueous solubility, in μmol/L. The corresponding statistical analysis of variance table is as follows:

Source	DF	Sum of squares	Mean square
Total	33	74.3	2.25
Regression	1	72.1	72.1
Residual	32	2.24	0.0699

$$R^2 = 0.970; F = 1030; \alpha = 0.005$$

Equation 1 allows an estimation within one order of magnitude of the partition coefficient of a given compound from its aqueous solubility.

The scatter of points in Figure 1 reflects the following two factors: temperature and analytical procedure. The temperature effect on the partition coefficient is generally about 0.01 log unit per degree near room temperature (20). The temperature effect on the solubility will be within the same order of magnitude. This corresponds to about a 25% change in either the partition coefficient or the aqueous solubility for every 10° variation in temperature. The maximum temperature effect would account for a deviation of no more than 50% or a factor of two in the plot. Varied analytical methods employed in the determination of the partition coefficient or the solubility account for the larger discrepancy in the literature values (16, 20, 29). Anomalies due to molecular structure can also be a possible cause. However, more information in this line is needed to make fair conclusions. In spite of the difficulty in analyzing some relatively unstable or insoluble chemicals, the experimental data in Figure 1 show an acceptable fit. It is of interest to note that such a correlation covers many classes of chemicals from hydrocarbons and organic halides to aromatic acids, pesticides, and PCB's. It also spans chemicals of different polarities (from nonpolar to polar) and of different molecular states (both liquid and solid).

The partition coefficient is defined in the form:

$$K = a_0/a_w \cong C_0/C_w \quad (2)$$

where *a*₀ and *a*_w refer to the activity, and *C*₀ and *C*_w to the concentration in the *n*-octanol and water phase. The close fit of the aromatic acids suggests that these compounds show no significant dissociation or dimerization in the system. The latter is in agreement with the findings of Fujita et al. (11) who reported an insignificant difference in the measured *K* values of benzoic acid with log *K* changing from 1.88 to 1.81 when the concentration in octanol phase varied from 10⁻² to 10⁻⁴ M. Similar findings were reported for phenoxyacetic acid and phenylacetic acid. Considering that the p*K*_a of these acids are in the neighborhood of 4, one would expect no more than a few percent dissociation in the aqueous phase. Obviously, it would be doubtful that Equation 1 would apply for salts, strong acids, and bases since the activities of these solutes in this case cannot be approximated by their concentrations.

The magnitude of the partition coefficients covered in Figure 1 ranges from less than 10² for unsubstituted aromatic acids to 10⁶ for PCB's and organochlorine pesticides. Hydrocarbons and organic halides fall in the range from about

Table I. Solubilities and Partition Coefficients of Various Compounds

Compound	Solubility in water, ppm	Log (<i>n</i> -octanol/H ₂ O partition coeff)
Benzene	820 (22 °C) ¹²	2.13 ¹¹
Toluene	470 (16°) ¹²	2.69 ¹¹
Fluorobenzene	1540 (30°) ¹²	2.27 ¹¹
Chlorobenzene	448 (30°) ¹²	2.84 ¹¹
Bromobenzene	446 (30°) ¹²	2.99 ¹¹
Iodobenzene	340 (30°) ¹²	3.25 ²⁰
<i>p</i> -Dichlorobenzene	79 (25°) ¹²	3.38 ¹⁹
Naphthalene	30 ¹²	3.37 ¹⁵
Diphenyl ether	21 (25°) ¹⁹	4.20 ¹⁹
Tetrachloroethylene	400 (25°) ¹⁹	2.60 ¹⁹
Chloroform	7950 (25°) ¹⁹	1.97 ¹⁴
Carbon tetrachloride	800 ¹²	2.64 ²²
<i>p,p'</i> -DDT	0.0031 ⁵ –0.0034 ⁴ (25°)	6.19 ²⁸
<i>p,p'</i> -DDE	0.040 (20°)	5.69 ²⁸
Benzoic acid	2700 (18°) ¹²	1.87 ¹¹
Salicylic acid	1800 (20°) ¹²	2.26 ¹⁴
Phenylacetic acid	16600 (20°) ¹²	1.41 ¹¹
Phenoxyacetic acid	12000 (10°) ¹²	1.26 ¹¹
2,4-D	890 (25°) ¹⁹	2.81 ¹¹
2,4,5,2',5'-PCB	0.010 (24°)	6.11
2,4,5,2',4',5'-PCB	0.00095 (24°)	6.72
4,4'-PCB	0.062 (20°)	5.58
Phosmet	25 (25°) ²³	2.83
Malathion	145 (20°) ²³	2.89
Fenitrothion	30 ²³	3.38
Dicaphon	6.25 (20°)	3.58
Parathion	24 ²³	3.81
Phosalone	2.15 (20°)	4.30
Methyl chlorpyrifos	4.76 (20°)	4.31
Dialifor	0.18 ¹⁷	4.69
Ronnel	1.08 (20°)	4.88
Chlorpyrifos	0.4 (23°) ⁷	5.11
Dichlofenthion	0.245 (25°) ³²	5.14
Leptophos	0.0047 (20°)	6.31

10^2 to about 2×10^3 and organophosphate pesticides from 10^3 to 10^5 . Leptophos shows an exceptionally high partition coefficient and low solubility in comparison with other organophosphates. Since the partition coefficient has been used in assessing the environmental and biological activities of the chemical, Equation 1 can provide a preliminary basis for a rapid evaluation of the probable behavior and fate of the chemical simply from its aqueous solubility, since this is generally more readily available and better established.

Equation 1 also will clarify some of the questionable partition coefficients reported for several chemicals. Although the aqueous solubility reported for DDT ranges from about 0.2 to 1000 ppb ($\mu\text{g/L}$) (2, 4, 5, 26, 30, 31), at this time it is accepted that the actual value is between 1 and 5 ppb. The published partition coefficient varies from about 9×10^3 to 1.6×10^6 (21, 28, 33). The accepted DDT solubility in the low ppb range would, according to Equation 1, predict a K value in the million range. The value of 1.6×10^6 calculated by O'Brien (28) is in good agreement with this estimate. Low K values have been reported for many compounds: 1 030 for aldrin, 13 560 for hexachlorobenzene (21), and 16 037 for 2,4,5,2',5'-PCB (25). According to the reported solubility of about 6 ppb for hexachlorobenzene (21), the authors would predict a K value of 1.5×10^6 which concurs with the value of 1.5×10^6 as calculated by Neely et al. (27). Based on the reported solubility values for 2,4,5,2',5'-PCB from 10 ppb of this laboratory to about 20 ppb of Metcalf et al. (25) and of Wallnofer et al. (34), the authors' experimental K value of 1.3×10^6 is in good agreement with that predicted from Equation 1.

As previously stated, the partition coefficient is related to physical adsorption on solids, biomagnification, and lipophilic storage. Equation 1 would extend those correlations to cover compounds using their aqueous solubilities without requiring the partition coefficient data.

The adsorption-solubility relationship may be understood by considering the (physical) adsorption of a chemical from aqueous solution. As adsorption is an exothermic process in which the adsorbate condenses from the bulk phase onto the adsorbed phase, an increase in solubility with increasing temperature would result in a reduction of the amount adsorbed. For compounds of comparable molecular size with different solubilities, the one with lower solubility, i.e., lower hydrophilicity, would tend to be adsorbed more favorably at a given equilibrium concentration, provided such adsorption is not complicated by other geometric effects (8, 9).

The correlation of biomagnification in mosquito fish (*Gambusia affinis*) with the aqueous solubilities of chemicals has been reported for some PCB's, DDT, DDE, aldrin, and many other organic compounds (21, 25). As an additional illustration, the authors are presenting a similar correlation in Figure 2 utilizing the reported biomagnification data of some selected organic chemicals in rainbow trout (*Salmo gairdneri*) (27). The solubilities of biphenyl and 2,4,2',4'-PCB in Figure 2 are obtained from Bohon and Claussen (35) and Wallnofer et al. (34), respectively.

The regression equation shows:

$$\log(\text{BF}) = 3.41 - 0.508 \log S \quad (3)$$

where BF = bioconcentration factor in rainbow trout and S = aqueous solubility in $\mu\text{mol/L}$. The associated statistical analysis of variance table is as follows:

Source	DF	Sum of squares	Mean square
Total	6	6.93	1.15
Regression	1	6.44	6.44
Residual	5	0.484	0.0968

$$R^2 = 0.930; F = 66.6; \alpha = 0.005$$

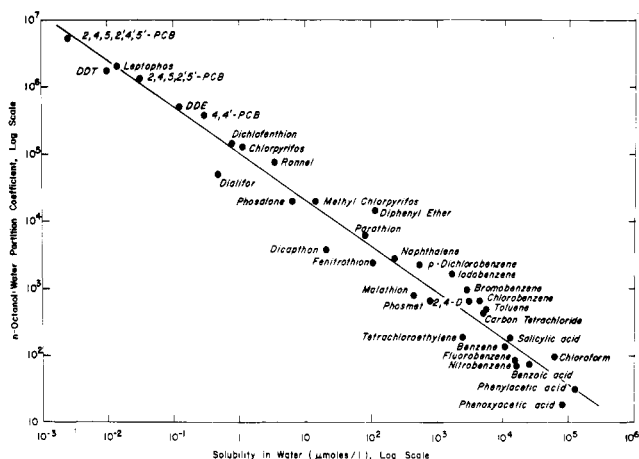


Figure 1. Partition coefficients and aqueous solubilities of various organic chemicals at room temperature

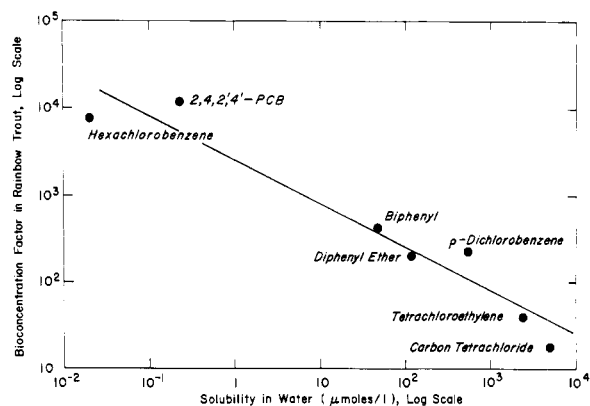


Figure 2. Aqueous solubilities and bioconcentration factors of organic chemicals in rainbow trout

The utility of the lipophilic storage-partition coefficient correlation has been demonstrated in the treatment of several cases of human poisoning involving the organophosphate dichlorofenthion (10). The relatively high partition coefficient for this compound provided an explanation of the anomalous symptomatology observed after ingestion. It was postulated that fat deposition with the resultant slow release into the circulatory system was the probable cause of the prolonged toxic manifestations. Leptophos, with a partition coefficient of more than an order of magnitude higher than dichlorofenthion, should exhibit similar behavior. An estimation of these partition coefficients from their available solubilities would be of value in understanding the unusual toxicological features encountered and the prescription of appropriate prolonged antidotal therapy.

An attempt has been made to illustrate a few interrelationships of the physical and chemical properties which guide the behavior and fate of chemicals in the environment. Hopefully, these correlations will prove useful for environmental scientists in assessing data and considering the possible effects of chemicals on the environment.

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Received for review June 14, 1976. Accepted November 29, 1976. Research supported by U.S. Public Health Service Grant ES-00040 from the National Institute of Environmental Health Sciences. Published with the approval of the Oregon State Agricultural Experiment Station as Technical Paper No. 4285.

Chlorination Reactions of Fulvic Acids in Natural Waters

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■ Chlorination of both drinking waters and wastewaters leads to the generation of various chlorinated degradation products of natural fulvic acids. Fulvic acids constitute the bulk of the organic matter in colored surface or ground waters. Metadihydroxybenzene structures are proposed as main reactive sites of the molecule. Experimental evidence is presented for possible reaction pathways leading to haloforms and other chlorinated by-products.

In 1974 Marx (1) gave particular notice to some disquieting effects on health as a result of chlorination of drinking water as observed in New Orleans by the group of Laseter.

Earlier in the year Harris and Page (2) had published statistical evidence for the carcinogenic properties of some micropollutants in drinking water from the Mississippi River. Some months later Dowty et al. (3) reported on their findings of 13 volatile halogenated organic compounds in tap water. At least five of these were detected in the blood plasma of local consumers of the water. Rook (4) in the meantime reported experimental evidence for the formation of haloforms as by-products of chlorination of naturally colored waters. Rook demonstrated that fulvic acids that impart the yellow color to the water are precursors for haloforms when treated with chlorine. Even traces of bromine generated by oxidation of the bromide ions in the water during chlorination produce analogous effects. Consequently, all colored water that is chlorinated, whether surface or ground water, must contain these

micropollutants, ironically introduced by a process to protect the consumers from waterborne diseases.

In 1975 the ubiquity of haloforms in chlorinated waters was duly confirmed by EPA's nationwide reconnaissance survey for halogenated organics (5). In all of the surveyed water treatment plants employing chlorine for disinfection, chloroform was present in the finished water. Positive correlations existed between haloform concentrations and the total organic matter content in the untreated water.

Fulvic Acid Structures

In the following report we shall give more evidence in support of our earlier experimental data obtained with resorcinol and aqueous peat extract as model substances for fulvic acids.

Since the classical studies of Oden, who in 1919 distinguished the group of yellow acids as a subgroup of humic acids, many researchers have contributed to the identification of the natural yellow acids in water environments with fulvic acids from the soil. Oden's classification is still generally used. It differentiates three groups according to solubility dependent on pH. The group that remains in solution at pH 1 is named fulvic acids. Fulvic acids are essentially small sized humic acid molecules.

Wilson (6) concluded from ultraviolet measurements that fulvic acids are the major component of organic color in surface waters. Black and Christman (7), in an extensive study covering 10 different sources of naturally colored waters, concentrated and fractionated the total amount of hydrophilic