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Biogeochemical Processes Governing Exposure and Uptake of Organic Pollutant Compounds in Aquatic Organisms

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This paper reviews current knowledge of biogeochemical cycles of pollutant organic chemicals in aquatic ecosystems with a focus on coastal ecosystems. There is a bias toward discussing chemical and geochemical aspects of biogeochemical cycles and an emphasis on hydrophobic organic compounds such as polynuclear aromatic hydrocarbons, polychlorinated biphenyls, and chlorinated organic compounds used as pesticides. The complexity of mixtures of pollutant organic compounds, their various modes of entering ecosystems, and their physical chemical forms are discussed. Important factors that influence bioavailability and disposition (e.g., organism-water partitioning, uptake via food, food web transfer) are reviewed. These factors include solubilities of chemicals; partitioning of chemicals between solid surfaces, colloids, and soluble phases; variables rates of sorption, desorption; and physiological status of organism. It appears that more emphasis on considering food as a source of uptake and bioaccumulation is important in benthic and epibenthic ecosystems when sediment-associated pollutants are a significant source of input to an aquatic ecosystem. Progress with mathematical models for exposure and uptake of contaminant chemicals is discussed briefly.

Introduction

Modern societies derive many benefits from mobilization of natural geochemical resources, such as oil, and from synthetic chemicals; for example, medicine, generation of usable energy, material for shelter, and chemicals for agriculture. The Organization for Economic Cooperation and Development (OECD) estimates that approximately 1000 to 1500 new substances enter daily use each year and add to the approximately 60,000 already in daily use (*1*), an estimate that is similar to that of the United States Council on Environmental Quality (*2*). Fortunately, only a small proportion of these chemicals pose threats to human health and living natural resources once they are released to the environment. This small proportion is still a large number. Butler estimated that 11,000 chemicals are manufactured in quantities that require assessment for environmental concern in that these quantities have the potential to pollute significant sectors of global ecosystems if released. In addition to these manufactured chemicals, there are chemicals of environmental concern generated by processes of treatment of industrial and domestic wastes by chlorination (*4–6*). The exact numbers are not important considerations in this paper. Rather, it is important that the assessment of toxicity and environmental fates and effects for these chemicals, and most important, assessment of human health risks associated with the presence of these chemicals in the environment, are substantial tasks requiring extensive knowledge of the behavior of these chemicals in the environment and their modes of toxic action. Studies of the past 15 to 20 years have provided evidence of the widespread distribution of recalcitrant synthetic organic chemicals such as the chlorinated pesticides and polychlorinated biphenyls (PCBs) in ecosystems worldwide even in locations far removed from sources of input. Aquatic ecosystems are not an exception, as has been well documented in numerous studies and reviews (*2,7,8*).

The presence of a xenobiotic compound in a segment of an aquatic ecosystem does not, by itself, indicate an adverse effect. Connections must be established between levels of exposure or tissue contamination and adverse effects. The combined understanding of the inputs, fates, and effects of chemical contaminants or pollutants in the environment has been termed ecotoxicology by groups of scientists (*3,9,11*). This paper focuses on a few advances in biogeochemical research concerned with the movement of organic chemicals through aquatic ecosystems and conditions of exposure and routes of up take by aquatic organisms. The focus is mainly on nonpolar, hydrophobic, medium molecular weight compounds such as PCBs, chlorinated pesticides, and aromatic compounds found in fossil fuels and their combustion products, chemicals often identified with the current concern about human health risks associated with chemical contaminants in fish and other aquatic species.

A stylized version of the biogeochemical cycle of a group of these compounds, polynuclear aromatic compounds (PAHs) is presented in Figure 1. It would be an immense task to quantitatively measure all the pathways and rates of reaction and movement for the many thousands of chemicals of concern through each of their individual biogeochemical cycles. Although individual chemical structure confers some specificity of environmental behavior and toxic action to each compound,

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there are principles by which we can group chemicals of similar structure and obtain some predictive capability of their biogeochemical behavior. This knowledge can then be used in concert with knowledge derived from effects studies to provide human health risk assessments and ecological risk assessments.

Physical Chemical Research

Fundamental properties related to the biogeochemical behavior of many organic chemicals of environmental concern have not been well characterized, and it is only in the past years that substantial progress has been made. For example, water solubilities are available for only relatively few compounds, especially for the medium to higher molecular weight nonpolar compounds such as PAHs, PCBs, and chlorinated pesticides. This situation is improving rapidly (*12–16*). The influence of temperature and salinity on solubility has been less extensively investigated (*16–19*) and requires much further research. Initial assessments of the influence of temperature and salinity on solubilities yield factors of 2 to 6 for medium to higher molecular weight range PAHs (e.g., phenanthrene to benzo[*a*]pyrene) over the range of temperatures and salinities normally found in coastal waters and the open ocean (*19*). However, the anomalous behavior of benzanthracene compared to other PAH tested suggests that important knowledge has yet to be gained for key aspects of factors influencing solubilities in seawater (*19*).

Calculation or estimation of water solubilities from theoretical and empirical considerations is an active area of research, and some progress has been made using molecular surface area and volume and activities of organic solutes in organic phase and activity coefficients in aqueous phase (*20–22*). The wide range of solubilities of these essentially hydrophobic compounds and the very low solubilities for compounds in the medium to higher molecular weight range, e.g., phenanthrene and 2,2',4,4',5,5'-hexachlorobiphenyl, are illustrated by the examples in Table 1.

Compounds with low solubilities have a tendency to sorb to surfaces, and this is another of the important physical-chemical parameters needed to understand the biogeochemical behavior of organic chemicals of environmental concern. Results from several investigations during the past 10 years have demonstrated

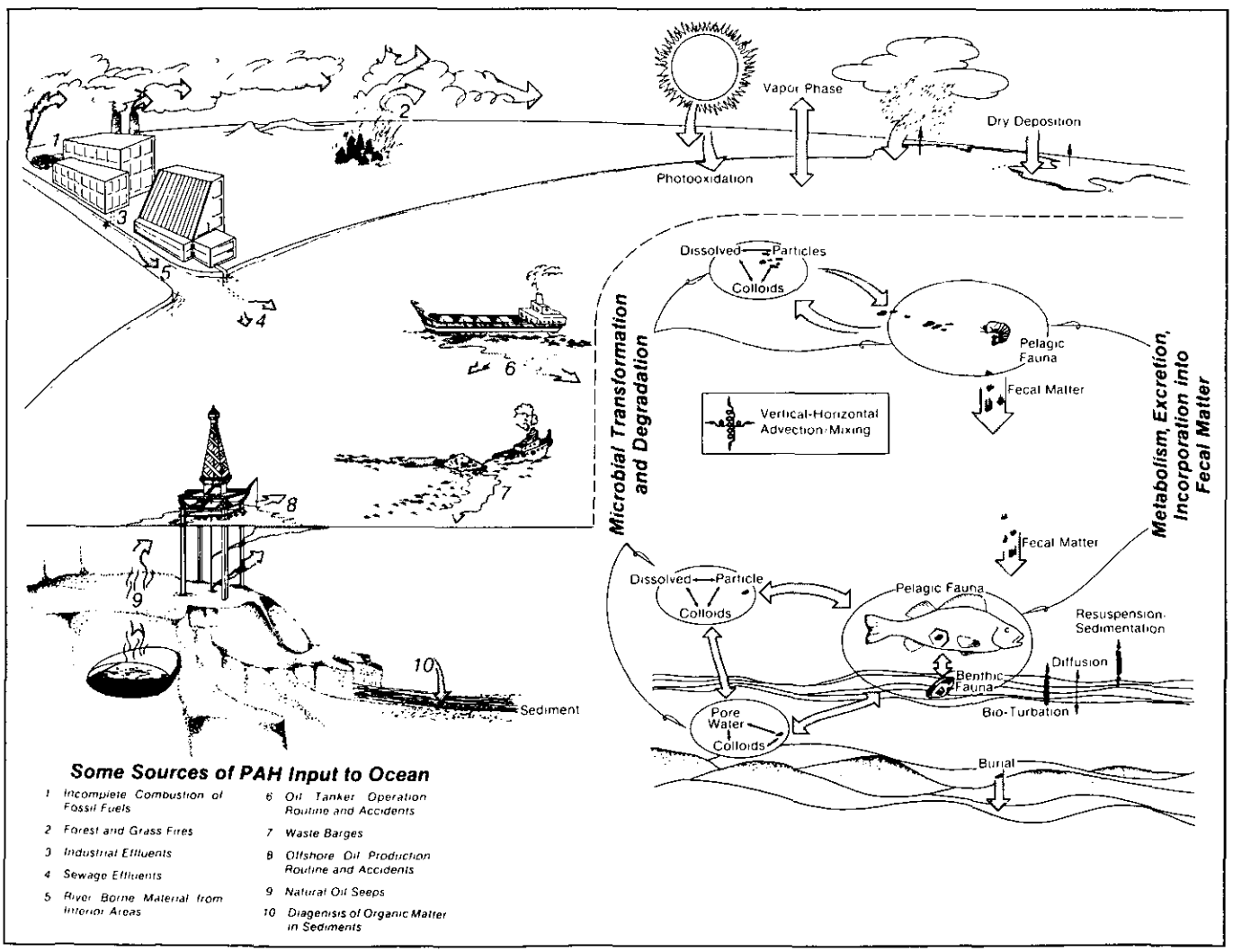

FIGURE 1. Biogeochemical cycle of PAHs in the ocean.

Table 1. Water solubilities of selected organic compounds at 25°C (*20*).

Compound	log S[a]
Acenaphthene	-3.86
Acrylonitrile	0.15
Aniline	-0.15
Benzene	-1.68
Benzo(*a*)pyrene	-6.26
Biphenyl	-3.88
1,3-Butadiene	-1.86
Carbon tetrachloride	-2.31
Chlorobenzene	-2.35
Dibenz(*ah*)anthracene	-6.28
m-Dichlorobenzidine	-3.01
3,3′-Dichlorobenzidine	-3.84
2,4′-Diclororbiphenyl	-5.32
1,2-Dichloroethane	-1.09
7,12-Dimethylbenzanthracene	-5.71
Ethylbenzene	-2.80
Hexachlorobenzene	-5.48
2,2′,4,4′,5,5′-Hexachlorobiphenyl	-7.66
3-Methycholanthrene	-6.54
Naphthalene	-3.04
Pentachlorobenzene	-4.85
Phenanthrene	-4.42
Styrene	-2.57
1,2,3,5-Tetrachlorobenzene	-4.33
2,2′,5,5′-Tetrachloroethane	-1.75
Toluene	-2.22
1,3,5-Trichlorobenzene	-4.09
1,1-Trichloroethane	-2.00

[a]S, molar solubility.

that sorption is mainly a simple partitioning of neutral, nonpolar hydrophobic compounds from the aqueous phase to a bulk nonaqueous phase (*23–26*). Organic constituents of natural sorbents are primarily responsible for sorption of hydrophobic organic compounds if the organic carbon content of the sorbent exceeds 0.1% (*27*). Thus, a partition coefficient K_p can be expressed in terms of the organic carbon content of the sorbent and in terms of a partition coefficient K_{oc} between water and a hypothetical sorbent of 100% organic carbon of a natural organic material:

$$K_p = f_{oc} \bullet K_{oc} \quad (1)$$

The partition coefficient K_{oc} can in turn be related to the octanol/water partition coefficient of a given compound by the following equation:

$$\log K_{oc} = a \log K_{ow} + b \quad (2)$$

where parameters a and b are empirically derived for different groups of compounds. Some of the values reported for these parameters in different studies are summarized in Table 2 to illustrate the type of variation to expect.

The K_{ow} octanol/water partition coefficient of a compound has proven to be a very useful parameter in predicting the environmental behavior of hydrophobic chemicals. There are means beyond the scope of this paper to derive the relationship between K_{ow} and solubility (*29–31*), as reviewed in Brownawell (*32*) and Lyman et al. (*33*), among others. Thus, it is possible to determine the K_{ow} of a given compound experimentally and then calculate its solubility and sorption in aqueous systems. Handbooks with some of these properties are now becoming available (*33*), and on-line computer systems for environmental data of this sort are also becoming available.

The caveat that must be kept foremost in the minds of environmental scientists is that data are available for very few of the compounds of environmental concern and the concepts have been tested in relatively few of the myriad geochemical milieu found in aqueous ecosystems. For example, there have been only a few studies of compounds such as amines, carboxylic acids, and phenols (*34,35*). These studies demonstrate that the simple partitioning model used to describe sorption of neutral, hydrophobic molecules is of limited applicability to compounds that are partially or fully ionizable at natural waters pH values. It is possible to account for the influence of ionization and assemble predictive relationships (*35–37*) but relatively few types of compounds have been studied. The influence of pH and Eh on the environmental behavior of ionizable organic pollutants needs much further research because many of the present and future problems with environmental fates and effects of compounds involve considerations of polluted sediments and estuarine and coastal waters (*38–40*) where substantial ranges of pH and Eh are encountered (*41,42*).

Natural waters can be partitioned into particulate and dissolved phases by operational means of filtration, and the definitions of dissolved and particulate are operational. The partition parameters discussed in the preceding paragraphs, K_p and K_{ow}, are based on the simplest partitioning process, two phases, and this works reasonably for many soil-water systems and sediment-water systems. Three lines of evidence led Brownawell (*32*) and Brownawell and Farrington (*43,44*) to hypothesize that at least a three-phase partitioning model is needed to model the distributions of many hydrophobic compounds in interstitial waters of sediments: *a*) experimental evidence showing sorption of hydrophobic organic compounds to colloidal organic matter (*45,46*); *b*) high dissolved organic carbon (DOC) concentrations

Table 2. Estimation of K_{oc} based on K_{ow}: $\log K_{oc} = a \log K_{ow} + b$ (*28,33*).

Coefficients correlation			Number of compounds	Range of $\log K_{ow}$	Type of chemicals
a	*b*	r^2			
0.544	1.377	0.74	45	-3.0–6.6	Primarily agricultural chemicals
1.00	-0.21	1.00	10	2.1–6.3	Polycyclic aromatic hydrocarbons (PAH)
0.937	-0.006	0.95	19	2.1–6.3	(PAH) triazines, nitroalines
1.029	-0.19	0.91	13	0.4–6.3	Herbicides, insecticides (*23*)
1	-0.317	0.98	13	1.6–6.5	heterocyclic aromatic compounds
0.72	0.49	0.95	13	2.6–4.7	Chlorinated hydrocarbons, alkylbenzenes
0.52	0.64	0.84	30	0.5–3.3	Substituted phenyl ureas and alky-*N*-phenyl carbamates

in the interstitial waters of marine sediments (*47–51*); and *c*) the fact that 50 to 90% of the interstitial water DOC is colloidal (*48,52,53*).

Field and laboratory evidence from several studies with PCBs has established that partitioning of hydrophobic compounds to colloids or very small particles must be considered in many natural water systems to account for the observed distributions of hydrophobic compounds in the dissolved and particulate fractions of water samples (*43,44,54,55*). Nomographs similar to those of Figure 2 (*32*) have been presented (*46,55,56*) and are useful in providing first order assessments of the physical chemical state of a given hydrophobic pollutant in coastal, estuarine, riverine, lacustrine, and interstitial waters.

The association of a significant proportion of the interstitial water hydrophobic compounds such as the PCBs with a colloid fraction has important implications for the assessment of the flux of these compounds across the sediment-water interface (*32*). First-principle arguments indicate that hydrophobic compound interactions with colloids will also be important in modeling the environmental behavior of compounds that enter estuarine waters across a salinity and particulate matter gradient as material is carried down river, or in the case of discharge of an industrial or municipal sewage effluent rich in organic matter content. Much of what is known about colloidal organic matter in natural waters is descriptive in nature (*51,57,58*). The relatively new findings and hypotheses about the importance of hydrophobic compound interactions with colloidal organic matter should add to previous observations and hypotheses concerning the importance of macromolecular organic matter and colloidal organic matter in contemporary biogeochemical cycles and stimulate new research efforts on colloids in natural water systems.

Biological Uptake, Release, Metabolism, and Distribution

Equilibrium Considerations

The concept of treating the relationship between chlorinated pesticides in aquatic organisms and their aqueous habitat as a pseudoequilibrium process was first proposed by Hamelink et al. (*59*). Stated in simple terms, exchange across membrane surfaces (e.g., gills) controls partitioning of a pollutant between the organism and water. The concept was shown to be consistent with data for DDT family compounds and PCBs in oceanic ecosystems (*8,39*) and was widely adapted for understanding of the relationship between hydrophobic pollutants such as PCBs, chlorinated pesticides, and PAHs in organisms and in waters of the organisms' habitat (*3,8,33,60*). Aquatic mammals and birds, being air breathing organisms, were exceptions to the concept.

Thus, direct uptake and release of organic pollutants from and to water joined food transfer and excretion as modes of input/output for pollutants in aquatic organisms and became the dominant modes in conceptual thinking and data interpretation. A procedure for assessing bioaccumulation potential under equilibrium conditions was needed, and Neeley et al. (*61*) were among the first to introduce the useful concept that K_{ow} could be used for this purpose. A compilation of data depicting the relationship between K_{ow} and K_b (bioaccumulation = concentration in organism/concentration in water) shows that the relationship

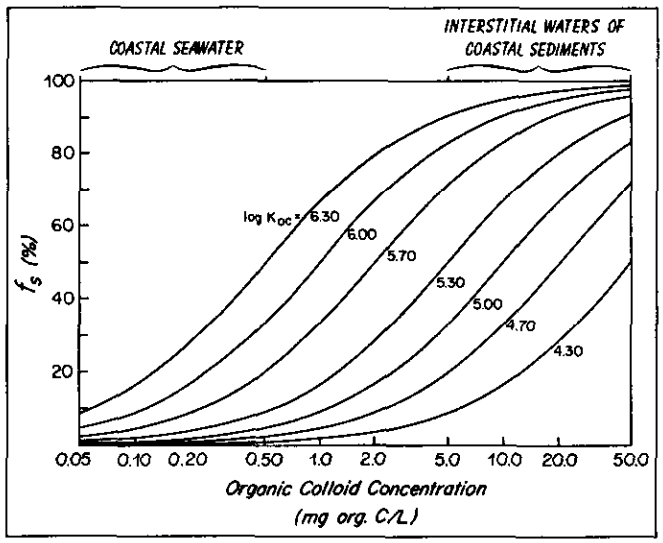

FIGURE 2. Nomograph for relationships between fraction (f_s) of a given hydrophobic compound sorbed to organic colloids, organic colloid concentration, and log K_{oc} of the compound. From Brownawell (*32*).

is predictive to a first approximation within a factor of 2 using Eq. (3) (*62*).

$$\log K_b = 1.00 \log K_{ow} - 1.32 \qquad (3)$$

There are also significant departures from the predictive relationship (*10,63,64*). These departures could be due to nonequilibrium conditions at the time of the measurements used to calculate K_b. Eq. (3) may not apply for hydrophobic compounds with $K_b > 6$, i.e., superlipophilic compounds (*62*). Connell and Miller (*65*) submit that the equation must be modified to take into account molecular shape, e.g., planar and nonplanar PCBs. Hawker and Connell (*63*) have noted that little attention has been paid to the time required to attain equilibrium, with the result that some of the data in the literature may underestimate K_b. There is also the problem of the physical form of the total of the organic chemical concentration present in the water. However, the influence of dissolved organic material in the water and colloidal organic matter cannot be ignored. It may well be that only that portion of the material in the water not associated with the organic matter is available for partitioning with the organism. Experiments with the uptake of nonionic hydrophobic organic compounds such as petroleum compounds and PAHs by organisms have demonstrated reduced uptake when DOC-humiclike material is present in comparison to aqueous systems with low dissolved organic matter (DOM) concentrations (*66,67*). Table 3 (*67*) illustrates this point.

Another important physical-chemical consideration is sequestering or entrapment of an organic pollutant within a particle. Equilibrium partitioning calculations for sediment-water interactions for the hydrophobic compounds using K_{ow}s may not be applicable for sequestered or entrapped compounds. Farrington et al. (*68,69*) noted that comparison of PAHs for bivalve mollusks, sediments, and polychaetes in sediments in the same general locations revealed substantial differences between sediments and the biota. The causative factor was hypothesized

Table 3. Bioconcentration factors observed after 30-hr exposure of *D. magna* to 3-methylcholanthrene in the presence of different concentrations of dissolved humic material.*

DHM concentration, mg C/L	Fraction of MC bound to DHM	Observed BCF (±SD)	Fractional decrease in BCF due to presence of DHM
0	0	13.209 (± 653)	—
0.15	0.05	12.171 (± 194)	0.07
1.5	0.32	8.121 (± 161)	0.38
15.0	0.82	2.311 (± 127)	0.82

*See McCarthy et al. (*67*) for details. Abbreviations: DHM, dissolved humic material; MC, 3-methylcholanthrene; BCF, bioconcentration factor.

to be different sources and physical chemical form for the PAHs. Two types of PAHs were thought to be present: those found principally sequestered or entrapped in soot and pyrolytic source particles but extractable with normal solvent extraction procedures; and those entering aquatic ecosystems in water accommodated form or sorbed in a relatively easily partitioned form with particulate matter, e.g., petroleum from spilled oil or waste oil in effluents. These observations and hypothesis are consistent with observation concerning physical-chemical speciation of PAHs in an estuarine ecosystem (*70*) and the recent data on bioavailability of PAHs in a coastal sediment (*71*). The preceding are but a few examples illustrating that the accurate application of the equilibrium partitioning concept or other approaches to estimating bioaccumulation of a pollutant organic compound from a concentration of the compound measured or estimated for an organism's habitat must take into account the physical-chemical form of the compound.

Kinetic Considerations

The need for more accurate, predictive capabilities beyond those of equilibrium considerations seems apparent. Hawker and Connell (*63*) and Connell (*72*) have offered an insightful review of the relationship between kinetic considerations and equilibrium considerations. Eqs. (4) to (7) are from their work:

$$dC_b/d_t = k_1C_w - k_2C_b \tag{4}$$

where C_b is concentration of pollutant in an organism; C_w is concentration of pollutant in the water phase; k_1 is a first-order uptake rate; k_2 is a first-order release rate or clearance rate from the organism; and t is time.

Water in the natural environment is a very large component relative to the organism, and thus C_w is assumed to be constant and not affected by bioaccumulation or release in this treatment of the equations. It is easily shown that at equilibrium

$$C_b/C_w = k_1/k_2 \tag{5}$$

$$\text{since } C_b/C_w = K_b, \textit{ then } K_b = k_1/k_2 \tag{6}$$

and substituting into Eq. (3), it can be shown that $k_1/k_2 = 0.048\ k_{ow}$. Thus, k_1 and k_2 are related to K_{ow} in the equations, and recent assessments of available data show that this is indeed the case (*63*). There is an inverse correlation between log $(1/k_2)$ and log K_{ow} ($r = 0.974$) and a direct correlation between log (k_1) and log K_{ow} ($r = 0.974$) up to log $K_{ow} = 7$. Equations of the form

$$\log K_1 = 0.337 \log K_{ow} - 0.373 \tag{7}$$

can be written and adequately describe most of the data in the literature. The fault with Eq. (7) is that it requires ever increasing k_1 with increasing K_{ow}, and it is not valid above log $K_{ow} = 6$, i.e., for the extremely hydrophobic or superlipophilic compounds. Hawker and Connell (*63*) submit that there must be an upper limit to k_1 in part based on the efficiency with which a compound can be transferred across membranes, which is in turn related to compound shape and size and in part the ventilation rate of organisms which must have an upper limit for tissue such as gills.

First-order rate kinetics are useful in explaining some of the data concerning uptake and release of pollutants. An example illustrates this point. Two small no. 2 fuel oil spills occurred in the Cape God Canal 3 years apart but at the same time of the year within 1 week of each other [nearly duplicate experiments (*73*)]. Biological half-lives derived from Eq. (8) and (9) were determined from data exemplified by Figure 3.

$$C_t = C_0e^{-kt} \tag{8}$$

where C_t is concentration in tissue at time t: and C_0 = concentration in tissue at time 0.

$$t_{bl/2} = \frac{0.693}{k_2} \tag{9}$$

where $t_{bl/2}$ is the biological half-life.

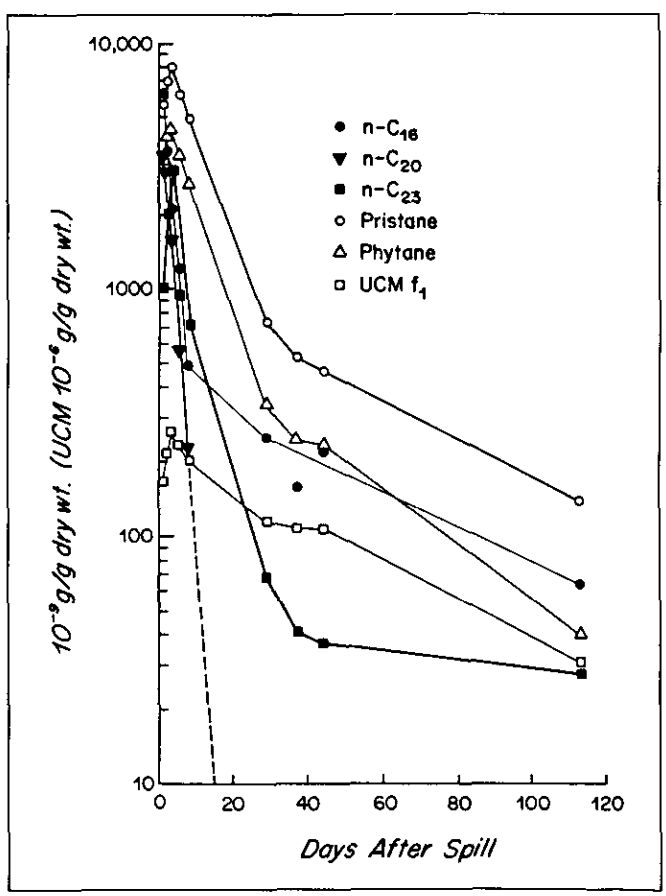

FIGURE 3. Time course of no. 2 fuel oil compounds released from *Mytilus edulis* after a contamination from a no. 2 fuel oil (*73*).

Table 4. Comparison of biological half-lives of no. 2 fuel oil compounds: 1978 and 1983 oil spill (73).

	$T_{b½}$, days	
	1978[a]	1983
Compounds	Day 1–21	Day 3–29
n-C_{16}	0.99	8.7
Pristane	7.7	6.3
Phytane	6.9	5.8
n-C_{23}	4.6	5.8
Σ C_2-naphthalenes	5.8	1.5
Σ C_3-naphthalenes	7.7	5.8
Phenanthrene	17	5.8
Σ C_1-phenanthrenes	9.9	6.9
Σ C_2-phenanthrenes	69[b]	9.9
Unresolved complex mixture f_1	14	17
Unresolved complex mixture f_2	14	35

[a]Recalculated for 1978 data. These are slightly different from those reported in Farrington et al. (75) because of an error in a calculator program discovered since that publication.
[b]Correlation fit of data to equation is poor.

A compilation of half-lives is given in Table 4. Review of the data for uptake and release of petroleum compounds, PAHs, and PCBs by bivalves indicates clearly that the relatively simple equations of the form of Eqs. (8) and (9) are applicable during the first 14 to 30 days . There are discernible departures from the semilog plots after 14 to 21 days in the example shown in Figure 3 depending on the compound considered. Concentrations of pollutant chemicals in the habitat and duration of exposure are known to be important considerations influencing rate of uptake and release of pollutants (60,77). For example, it has been suggested that a combination of high exposure concentrations and longer time of exposure, as would be expected in urban harbor areas, result in much slower overall release of pollutant hydrocarbons when bivalve mollusks are moved to cleaner waters.

The Stegeman and Teal (78) multiple-compartment model approach to explaining data from experiments and field observations derives from the fact that there are many tissue types in bivalves. It is not difficult to visualize this model, and it is supported by several studies (60,73,75,79). An example would be a simple three-compartment model bivalve of gills, circulatory fluid, and energy storage reserve lipids. Initial uptake across the gills is rapid, followed by slightly less rapid transfer to circulatory fluid, followed by much slower transfer to, and accumulation in, storage lipid reserves. Long-term exposure would result in accumulation of pollutant in lipid energy reserves until equilibrium or saturation of the storage capacity is attained. Transfer of the bivalve to clean water or removal of the pollutant from the water of its habitat reverses the process. Exchange of the pollutant from gill tissue to water is rapid, mobilization of the pollutant in the energy storage lipid is slower, and this accounts for an initial rapid release followed by much slower release in the longer term. Equations for assessment of multiple compartment models have been described by Mackay and Hughes (80) and involve steady-state assumptions in all but the target compartment, i.e., the compartment for which the flux of compound is of interest. Further elaboration of those models is beyond the scope of this synopsis.

It is well established that physiological status of organisms such as spawning, postspawning, prespawning, temperature of the habitat, food availability, as well as exposure concentration and duration, influence the uptake, retention, and release of pollutant organic chemicals by bivalve mollusks (60,77,79). Metabolism of PAHs and xenobiotics such as PCBs is thought to be much less active for bivalves than for fish, crustacea, or polychaetes (40,81). However, there are reports that bivalve tissues contain enzymatic activity related to metabolism of PAHs, and perhaps similar compounds (82,83). This is consistent with data showing isomer-specific changes in relative abundance of C_2 and C_3-alkylated phenanthrenes in the latter stages of release of no. 2 fuel oil compounds by mussels contaminated by an oil spill (73).

A fallacy of the use of first-order kinetics in this situation is readily apparent if we derive Eq. (10) from Eq. (4).

$$C_b = (k_1/k_2)\, C_w\, (1\text{-}e^{-k_2 t}) \qquad (10)$$

Theoretically, equilibrium can only be attained after infinite time. It is useful to circumvent this problem and work with the concept of a close approach to equilibrium, e.g., 0.99 of value, as has been used by Hawker and Connell (63). They have then derived an equation that relates time to equilibrium to K_{ow}:

$$\log t_{eq} = 0.663 \log K_{ow} - 0.284 \qquad (11)$$

This allowed them to predict that with compounds with log $K_{ow} < 6$ attain equilibrium within 1 year. Compounds for which log $K_{ow} > 6$ require too much time to approach equilibrium for reasonable measurements to be made. Hawker and Connell (63) also proposed a term t_s, as time to significant bioaccumulation with significant bioaccumulation being 1% of the bioaccumulation at equilibrium. They then used an equation relating t_s to log K_{ow} to calculate that compounds with a log K_{ow} of 10 are bioaccumulated significantly only after a minimum of 0.5 years. Aquatic organisms bioaccumulate a significant amount, i.e., 1% of equilibrium bioaccumulation concentration, of compounds with log K_{ow} of 13 after a minimum of 50 years. The latter time exceeds the lifetime of most aquatic organisms of concern, and the former time exceeds seasonality in temperate zones. The physiological status and other factors described previously that influence concentrations of organic chemical pollutants in an organism will change conditions of the organism or habitat well within the time required to reach significant bioaccumulation for hydrophobic pollutants with log K_{ow} of approximately 8 or 9 or greater. This limits the predictive capabilities of the kinetic equations described above.

Metabolism

Organisms other than bivalves are capable of reducing concentrations of hydrophobic organic chemical pollutants by exchange with water. In fact, some of the initial equilibrium partitioning work related to fish. In addition, fish, crustacea, and polychaetes have active enzyme systems capable of metabolizing substantial portions of bioaccumulated PAHs, some PCBs, and similar compounds (39,40,71,81). Research concerned with metabolism of organic chemical pollutants has expanded greatly in the past decade, and much detailed information is becoming available. The scope of this paper cannot provide many details. It suffices to state the obvious: concentrations in tissues and distributions among excretion of metabolites can be a function of a variety of tissues and conditions controlling enzymatic activity. These

conditions include spawning, nutritional status, conditions and duration of exposure to organic pollutants, mix of pollutants, and life cycle stage of the animal.

Exposure to xenobiotic organic compounds or PAHs has been shown to induce activity of mixed-function oxidases capable of metabolizing these xenobiotics. Extent of enzyme activity can be species-specific even for related species. Reichert et al. (*84*) exposed two species of deposit feeding amphipods to sediment-associated radioactively labeled benzo(*a*)pyrene (BaP). One species converted a higher proportion to metabolites, even though both species bioaccumulated the compound. The explicit lesson is that there are drawbacks to mathematical modeling of uptake, retention, metabolism, and release by extrapolating data from one species to another. This must be kept in mind when pragmatic approaches to the problems of a complex environment with thousands of species forces such extrapolations.

Another important example involves interactive effects of one chemical pollutant on another. Stein et al. (*85*) have shown that benzo(*a*)pyrene and PCBs interact and influence the extent of uptake and metabolism of each. Simultaneous exposure of English sole (*Paraophyrus vetulus*) to radioactively labeled BaP and PCBs increased concentrations of BaP-derived metabolites in the whole fish and decreased concentrations of PCBs and metabolites in some tissues and bile relative to results from separate exposure to radioactively labeled compounds in sediment-associated form.

Transfer in Food As a Source of Organic Pollutants

The water-organism partition hypothesis of Hamelink et al. (*59*) contributed significantly to the understanding of aquatic pollution and bioaccumulation and has prevailed for 15 years (*39,40,86*). This hypothesis evolved from experimental evidence in the laboratory and explained observations in the field obtained during the late 1960s and early 1970s when there were significant discharges of pollutant organic chemicals via effluents and transfer to water in aquatic ecosystems from atmospheric transport and runoff from land.

Several decades of input have resulted in accumulation of some compounds such as PAHs and PCBs and chlorinated pesticides in sediments (*9,39,40,87,88*). Several researchers have demonstrated that organisms living in or on polluted sediments can bioaccumulate the pollutants (*71,88–91*). Almost all experimental designs or field observations that were reviewed by these authors did not allow a sorting out of the relative importance of water-organism partitioning and sediment ingestion or water-organism partitioning and food ingestion in terms of contributions to bioaccumulated pollutant. In many cases, release of the pollutant from sediment by desorption was thought to cause elevated concentrations in the water followed by organism-water partitioning to achieve bioaccumulation. Oliver (*92*) has presented estimates (Table 5) for Lake Ontario that loadings of chlorinated hydrocarbons by desorption from sediments are in the same order of magnitude as are Niagara River inputs.

An elegant experiment by Rubinstein et al. (*89*) involving a multiphase experimental exposure design of a demersal fish (*Leiostomus xanthurus*) feeding on a polychaete (*Nereis virens*) with both fish and polychaete exposed to PCB-contaminated sediments has been completed. Rubenstein et al. conclusively demonstrated the importance of dietary input of PCBs from the polychaetes that had, in turn, been contaminated by PCBs originally in the sediment.

Table 5. Comparision of contaminant loadings to Lake Ontario from bottom sediment desorption and Niagara River discharge (kg/y) (*92*).

Chemical[a]	Sediment desorption	Niagara River	Chemical	Sediment desorption	Niagara River
1,3-DCB	3	1600	2,4,5-TCT	4	190
1,4-DCB	11	5800	2,3,6-TCT	1	120
1,2-DCB	2	2600	PCT	6	50
1,3,4-TCB	9	140	HCBD	6	130
1,2,4-TCB	19	2400	OCS	3	10
1,2,3-TCB	2	540	PCB 18	1	70
1,2,4,5-TeCB	12	340	PCB 52	4	120
1,2,3,4-TeCB	7	760	PCB 40	1	16
QCB	7	240	PCB 101	4	90
HCB	19	120	PCB 153	4	60

[a]Abbreviations: DCB, dichlorobenzene; TCB, trichlorobenzene; TeCB, tetrachlorobenzene; QCB, pentachlorobenzene; HCB, hexachlorobenzene. TCT, trichlorotoluene; PCT, pentachlorotoulene; HCBD, hexachlorobutadiene; OCS, octachlorostyrene; PCB, polychlorobiphenyl.

These results have very important implications for future efforts at modeling biogeochemical behavior, bioaccumulation, and, indeed, risk assessment for hydrophobic organic chemical pollutants in aquatic ecosystems. If a primary source of input to an aquatic ecosystem is slow release from sediments that have been polluted with inputs from various sources that have been reduced or eliminated in more recent times, then it is plausible that uptake by benthic organisms such as small bivalves, polychaetes, and crustacea followed by predation by larger organisms such as fish may be a significant source of bioaccumulated chemicals for the larger organism. In essence, food web transfer among benthic and epibenthic species may be as important as organism-water partitioning when aquatic ecosystems switch from top down (from effluent and atmospheric inputs) to bottom up (releases or transfers from polluted sediments) as regulatory controls reduce the top down sources of input. Even for aquatic ecosystems where inputs continue from effluents, the atmosphere, and runoff, there may be some ecosystems where a pseudo-steady-state approximation exists or is being approached, and food web transfer is now important for some species.

Mathematical Models of Coastal Ecosystems: From Inputs to Concentrations in Edible Tissues of Marine Organisms

It is now possible to incorporate the knowledge and predictive capabilities reviewed in earlier sections of this paper into dynamic models of entire ecosystems involving geophysical fluid dynamics, turbulence, sediment transport, and life-cycle stages of various species and arrive at some predictive capabilities with respect to contamination of edible tissues from marine organisms. O'Connor et al. (*93*) and Spaulding et al. (*94*) among others have presented such models.

One example suffices for the purpose of this paper. O'Connor et al. (*95*) have presented a mathematical model for the distribution and movement of the chlorinated pesticide, kepone, in the James River Estuary. Figure 4 shows the food chain portion of

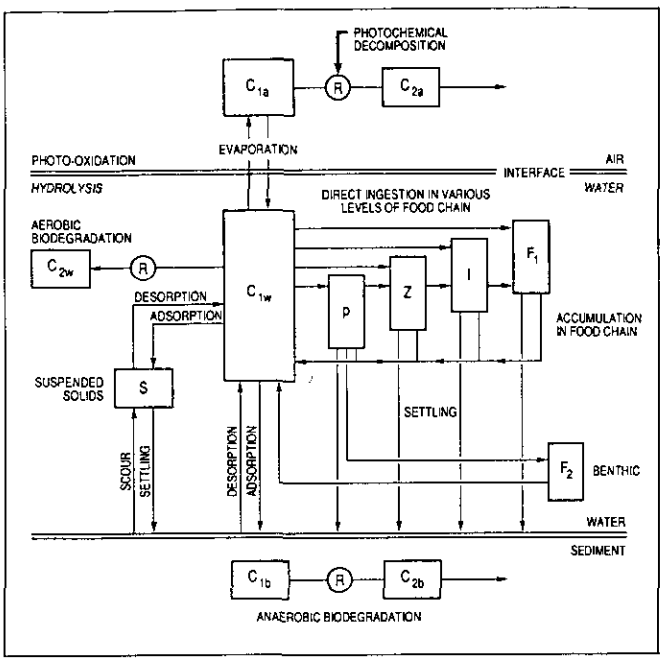

FIGURE 4. Kinetic and transfer routes of toxic chemicals in the water, bed, and food chain. From O'Connor and Piazza (9).

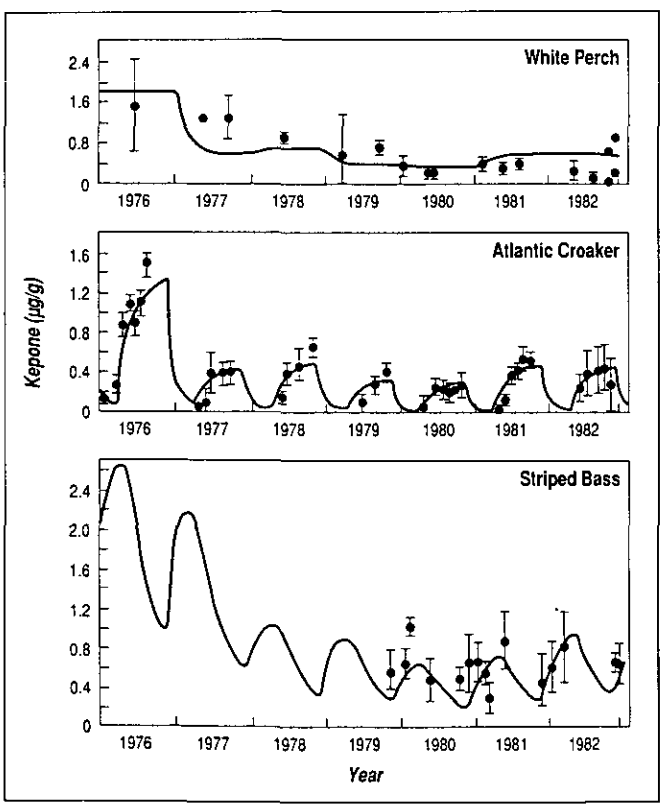

FIGURE 5. Comparison of observed and model calculated kepone concentrations in Atlantic croaker, white perch, and striped bass in the James River, Chesapeake Bay example. From O'Connor and Piazza (93).

the model. The comparisons between the calculated kepone concentrations and the measured kepone concentrations in white perch, Atlantic croacker, and striped bass are given in Figure 5. The agreement seems reasonable to a first approximation although reasonable agreement depends on the use of the model and the degree of uncertainty that will be accepted, and this probably will vary depending on the perceived or real importance of relative risks to aquatic biota or human health. The types of mathematical models exemplified by the work of O'Connor et al. (*93,95*), Thomann (*96*), Spaulding et al. (*94*), and references cited therein are becoming an essential part of environinental risk assessment for issues ranging from oil spills to remedial action plans for Superfund sites in coastal estuaries (*94,97*).

General Discussion

There has been substantial progress during the 1970s and 1980s in all aspects of biogeochemical research related to the issues of bioavailability and disposition of toxic organic chemicals: solubility, sorption, uptake, metabolism, retention, release/excretion. Predictive equations have been derived or have evolved empirically that tie molecular structural characteristics or properties to biogeochemical behavior. Coupling of the portions of biogeochemical models that deal with transfer back to people via consumption of living aquatic resources and potential impacts on human populations, e.g., consumption of carcinogens in edible portions of fish tissues (*98*), presents an important and powerful tool for realistic regulation of pollution for the protection of human health.

However, caution must be exercised that the elegance and complexity of a series of coupled mathematical equations does not evoke a false sense that accurate predictive capabilities of wide-ranging applicability are a proven reality in either the scientific community or in the policy, regulation, and management communities concerned with aquatic pollution problems. Thus far, the concepts and hypotheses reviewed briefly in this paper have been tested on relatively few chemicals and relatively few biota and ecosystems and, with few exceptions, for relatively short periods of time of days to months. Reuber et al. (*99*), in the closing statement of a recent paper concerned with chemical equilibria and transport at the sediment-water interface, stated, "Finally, models as described here will remain of limited value until they can be applied and validated in real situations." Nevertheless, an optimistic view is in order because research is gaining on the problems, and the science has evolved from mainly a descriptive endeavor to quantitative approaches involving a mature healthy mix of theory, experimentation, field observation, research, and monitoring.

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