

1988

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Robert C. Hale
Virginia Institute of Marine Science

Robert J. Huggett
Virginia Institute of Marine Science

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August 1988

Understanding the Estuary: Advances in Chesapeake Bay Research

Proceedings of a Conference
March 29-31, 1988



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**Proceedings of a Conference
29-31 March 1988
Baltimore, Maryland**

**Maurice P. Lynch and Elizabeth C. Krome
Editors**

August 1988

**CRC Publication No. 129
CBP/TRS 24/88**

Bioavailability of Organic Pollutants to Aquatic Organisms

Robert C. Hale and Robert J. Huggett

Division of Chemistry and Toxicology

Virginia Institute of Marine Science

College of William and Mary

Gloucester Point, Virginia 23062

INTRODUCTION

Settlement of the Chesapeake Bay region began in earnest in the early 17th century. The native American population and early colonists were impressed by the abundance of fish and shellfish and located their population centers to take advantage of these and other natural resources. Introduction of wastes into the bay was coincident with this settlement. As the human population increased, so did the pressure on the ecological system. In the 20th century significant quantities of synthetic chemicals began to be introduced, many of which were toxic and nonbiodegradable [Faust and Hunter 1971]. Today the areas surrounding the bay are experiencing unprecedented development; introduction of toxic organic pollutants has correspondingly increased. Water quality and the abundance of many aquatic organisms have suffered accordingly [O'Connor and Huggett 1988].

The bioavailability of organic pollutants is of recent concern and has not been as thoroughly studied as that of toxic metals. The initial belief was that "insoluble" organic xenobiotics were not available to aquatic organisms. They were assumed to be eliminated by irreversible binding to bottom sediments, which supposedly removed any significant threat to the ecosystem. More recently, we have come to realize that the term insoluble is a misnomer. All organic compounds possess some water solubility. In fact, it has been established that many low-solubility compounds are bioaccumulated or biomagnified to high concentrations in the tissues of organisms. Within several homologous series of organic compounds, toxicity has been negatively correlated with water solubility [Veith et al. 1983; Konemann 1981]. However, since chemically dissimilar compounds may exert their toxic effects via different mechanisms and influence different physiological functions, the solubility/toxicity relationship may not be directly applicable across series; the case of alcohols [Veith et al. 1983] versus organo-

phosphorus pesticides [DeBruin 1976] is a good example. Sediments in many cases have been determined to be a source of toxic compounds, rather than merely a sink [Willford et al. 1987]. As a consequence, we have been forced to re-evaluate our understanding of the bioavailability of organic pollutants and their significance to the health of the Chesapeake Bay ecosystem.

PHYSICAL, CHEMICAL, AND BIOLOGICAL BASIS OF BIOAVAILABILITY

Water

Water is an obvious and important route for the exposure of aquatic organisms to organic pollutants. The water solubility of a compound has a profound influence on its environmental fate and bioavailability. Biota may come into contact with high concentrations of compounds that exhibit significant solubility, e.g., alcohols, phenols, and benzenes. Accidental spills and untreated effluents may result in toxic concentrations of these compounds in the water, causing obvious acute effects such as fish kills.

Some of the most toxic organic compounds possess low water solubilities. Classes of these compounds detected in the Chesapeake Bay include polynuclear aromatic hydrocarbons (PAHs), heterocyclic aromatic compounds (HACs), and halogenated pesticides. Table 1 gives several specific examples. These compounds may be taken up by biota directly from water, although dissolved concentrations will generally be very low.

The mechanism of accumulation is believed to be simple partitioning from water into lipid-rich biological tissues [Esser 1986]. Active biologically mediated transport mechanisms are thought to be less prevalent for organic pollutants than for trace metals; many of the metals have critical functions in enzyme systems. The hydrophobic nature of lipophilic xenobiotics provides

the driving force for the partitioning process. The presence of nonpolar organic solutes is not compatible within the polar water phase.

Since laboratory experiments designed to determine bioavailability directly using fish and invertebrates are expensive and time-consuming, surrogate tests have been proposed. For example, the bioaccumulation tendency of a lipophilic organic generally correlates with the n-octanol/water partition coefficient or K_{ow} . Octanol has been suggested as a substitute for biological lipids in these experiments, although some researchers have suggested that critical differences exist in the thermodynamics of partitioning between water, fish lipids, and octanol [Oppenhuizen et al. 1988]. The classical approach for the determination of an octanol/water partition coefficient involves the addition of the test compound to a vessel containing mutually saturated octanol and water phases. The contents of the vessel are then thoroughly mixed and the system is allowed to come to equilibrium. The concentrations of the test compound in each of the two phases are subsequently determined and the coefficient calculated [Karickhoff and Brown 1979]. Difficulties (e.g., emulsions, detection limits, and contamination) are inherent in determinations for compounds possessing K_{ow} values greater than 100,000. Indirect measurement of the K_{ow} has also been suggested. For example, high-performance liquid chromatography (HPLC) has been used [Brooke et al. 1986]. Basically, a series of chemically similar compounds, for which the partition coefficients are known, are co-injected with the compound of interest onto a reverse-phase HPLC column. A correlation of retention time with K_{ow} is then determined and the partition coefficient for the compound of interest is calculated from this relationship. K_{ow} values are often expressed as logarithms, due to the magnitude of the values and the relationship of this parameter to bioconcentration factors (BCFs). Table 2 lists the log BCF and log K_{ow} of several organic compounds which have been detected in the environment. Note that the BCFs are less than the K_{ow} values.

Equations relating K_{ow} directly to bioconcentration factors (BCF) in various types of organisms and to water solubility have been reported [Isnard and Lambert 1988; Esser 1986]. It has been reported that concentrations of lipophilic pollutants will be similar in aquatic organisms in general, provided exposure has been equal, equilibrium has been established, and the relative lipid contents of the biota have been normalized [Adams 1988]. Obviously, differences in the biotransformation capabilities of the organisms may alter this relationship.

K_{ow} determinations provide no information concerning biotransformation or biological effects. These

Table 1. Water solubility [May et al. 1978] and LC50 [Trucco et al. 1983] for some common environmental contaminants. Values for LC50 encompass a variety of organisms and conditions and are used for illustrative purposes only.

	Compound (mg/l)	Solubility LC50 (mg/l)
Naphthalene	31.7	1.00
Ronnel	1.08*	0.49†
Phenanthrene	1.00	0.10
Benz(a)anthracene	0.009	0.01
Arochlor 1254	0.012*	0.003*
p,p'-DDT	0.003*	0.0002†

* Data from Chiou et al. (1977).

† Data from Johnson et al. (1980).

+ Data from National Research Council (1979).

phenomena are often significant. For example, English sole exposed to benzo(a)pyrene and Arochlor 1254 exhibited progressive accumulation of PCBs, but little accumulation of the PAH. This difference was attributed to extensive metabolism of benzo(a)pyrene by the fish [Malins et al. 1987]. Information on metabolism is very important in assessing the fate and effects of chemicals, especially since biotransformation may result in the production of more toxic or mutagenic products [Buhler and Williams 1988]. Bruggeman et al. [1984] observed that guppies bioaccumulated hexachlorobiphenyl, but not hexabromobenzene during aqueous exposures. They attributed this observation to the existence of an upper molecular size threshold, limiting transport across membranes. The importance of molecular volume has also been suggested by other workers [Doucette and Andren 1987]. Thus the use of octanol/water partition coefficients alone may result in

Table 2. The log BCF and log K_{ow} of several common environmental contaminants are given. Values are from Veith et al. [1979], except as noted.

Compound	log BCF	log K_{ow}
Naphthalene	2.63	3.37*
Pentachlorophenol	2.89	5.01
Phenanthrene	3.42	4.46
p,p'-DDT	4.47	5.75
Chlordane	4.58	6.00
Arochlor 1254	5.00	6.47

*Chiou et al. (1977).

an overestimation of the BCF in the case of extremely large molecules.

The major avenue of entry into biota for lipophilic compounds varies depending on the organism's size, morphology, and ecology [Knezovich and Harrison 1987]. For example, sorption to the general cell surface may predominate for single-cell biota, such as algae or heterotrophic bacteria. In higher organisms the body surface may contribute, but specialized respiratory structures (e.g., gills in fish) often are more important. Because of the large amounts of water processed by gills and the high lipid/water partition values (often >1,000,000), significant amounts of xenobiotics may rapidly bioaccumulate. Physiological changes may alter the bioavailability and thus the toxicity of organics to organisms. Conklin and Rao [1978] exposed molting adult grass shrimp to pentachlorophenol during various phases of the ecdysial cycle. They observed greater sensitivity and enhanced uptake immediately after ecdysis and attributed these effects to increased permeability of the cuticle. Newly molted blue crabs were observed to contain higher burdens of radiolabeled benzo(a)pyrene-derived material, compared with nonmolting crabs [Hale 1988] after laboratory exposure of these organisms.

Sediments

As previously mentioned, many organic compounds are rapidly sorbed to particulate matter. These compounds may adsorb to the mineral surface directly or to organic constituents of particles. The latter site is thought to be the most important [Hodson and Williams 1988]. Means et al. [1980], in determinations of batch equilibrium sorption isotherms, reported that adsorption of PAHs and HACs onto suspended soil/sediment was independent of substrate pH, cation exchange capacity, textural composition, or clay mineralogy. This statement is probably an oversimplification for direct application to waters of the Chesapeake Bay, because of the effects of factors such as pH on dissolved organics that may sorb lipophilic pollutants. These parameters are quite important in the behavior of polar organics and heavy metals. Grain size has been mentioned as a significant factor by some researchers [Marcus et al. 1988]. However, sediment grain size and organic content are generally correlated in the aquatic environment. The relationship of pollutant adsorption to organic content of sediments is well documented [Karickhoff and Brown 1979]. Equations relating sediment adsorption have been reported and generally employ coefficients such as K_{ow} [Dzombak and Luthy 1984].

The exact physical/chemical mechanism of sorption of chemicals to particulates is still uncertain. It has been postulated that the reduction of the water-organic

interfacial area achieved by sorption is critical [Mackay and Powers 1987]. The mechanisms of uptake into biological lipids and the organic constituents of particulates appear similar. In comparison, parameters such as hydrogen bonding and gross electrostatic attractions contribute little to the sorption of nonpolar organics to sediments [Voice and Weber 1983].

Sorption of organic pollutants reduces but does not eliminate availability to biota. Malins et al. [1987] reported significant correlations of the occurrence of liver disease in bottom-dwelling fish with the level of sediment PAH contamination. Huggett et al. [1987] found a variety of abnormalities in fish of the Elizabeth River, Virginia, compared with specimens from less polluted rivers. The Elizabeth is heavily contaminated with PAHs. Hargis et al. [1984] reported acute toxicity and lesion formation in fish held over Elizabeth River sediments in the laboratory. Fish exposed to water alone that had passed over these contaminated sediments also showed signs of chemical stress. This result indicated that the xenobiotics were quite bioavailable.

Duration of contact between lipophilic compounds and sediments has been observed to affect their rate of desorption and bioavailability [Varanasi et al. 1985; Voice and Weber 1983]. Similar observations in regard to the elimination of lipophilic residues from aquatic biota, i.e., biphasic depuration patterns, have been reported [Spacie and Hamelink 1982]. Haddock et al. [1983] reported that extraction efficiency of PAHs, using organic solvents, was a function of PAH/sediment contact time. Coal is an extreme illustration of the bioavailability question. It contains a large variety and quantity of PAHs. However, these compounds were found to be biologically unavailable to oysters during laboratory exposures to suspended coal dust [Bender et al. 1987]. Indeed, coal and activated charcoal are used in water treatment for the removal of dissolved organics. An analogous example may be the low availability of metals incorporated in the crystalline lattice structure of mineral grains. The transfer of lipophilic xenobiotics from organic reservoirs of sediments to those in biota does not present the favorable thermodynamics observed for partitioning from water to sediments or biota. Indeed, transfer from sediments to biota may entail movement into the polar water phase as an intermediate step. The octanol/water partition coefficient will describe the tendency for this movement to occur, desorption obviously being less favorable than sorption. Wood et al. [1987] observed that lower chlorinated congeners were preferentially desorbed from PCB-contaminated sediments. They also observed that dipteran larvae present in these sediments bioaccumulated PCB congeners containing two to four chlorines in preference to higher chlorinated congeners.

More typically, the bioconcentration of PCBs has been highest for congeners possessing five to seven chlorines, in accordance with their K_{ow} .

Evidence that accumulation and toxicity of organic xenobiotics are related to the positions of the substituents is also available [Kuehl et al. 1987]. Sediments collected from sites in the Hampton Roads area of Virginia were analyzed by capillary gas chromatography and a halogen-selective Hall detector [Hale, in preparation]. Significant concentrations of both PCBs and highly chlorinated polychlorinated terphenyls (PCTs) were identified at one location. Clams collected at this same site exhibited only PCBs, but the accumulation of lower-chlorinated PCT congeners observed in bivalves at another site indicated that PCTs were bioavailable.

Exposure of biota to particulate-associated xenobiotics may occur via direct epidermal contact with sediment particles. Contact with interstitial water, which contains higher concentrations of pollutants than the overlying waters, may be even more important [Knezovich and Harrison 1987]. Suspension feeders such as oysters may filter contaminated particles from the water, and other organisms (e.g., benthic worms) may ingest bulk sediment [Reynoldson 1987]. Again, bioavailability of this adsorbed material is lower than that of xenobiotics dissolved in water.

Resuspension of contaminated sediments via dredging operations has been observed to increase the bioavailability of Kepone in the James River [Lunsford et al. 1987]. Severe hydrographic events, e.g. floods and hurricanes, may resuspend sediments [Wood et al. 1987]. Bioturbation has also been reported to increase bioavailability of organic toxics. Reynoldson [1987] reviewed the results of several studies of bioturbation. He reported that tubificid worms transported 90% of hexachlorobenzene, pentachlorobenzene, and trifluralin to the sediment surface from a uniformly mixed sediment in laboratory experiments over a 50-day period. This resulted in a four- to six-fold increase in contaminants in the overlying water, compared with unperturbed systems. This work was done by Karickhoff and Morris.

Estuarine circulation and chemistry may result in the formation of turbidity maxima. This phenomenon is observed when suspended particulates flocculate upon contact with saline water and settle. The particles are then transported up-estuary by tidal currents to less saline water, where they may dissociate and again be carried downstream. This cycling leads to the development of a turbid zone with a high sedimentation rate. Lipophilic pollutants sorb to particulates and may concentrate in these zones, as was observed for Kepone in the James River [Huggett et al. 1980].

Surface Microlayer

Organisms on the surface of the water may be exposed to elevated concentrations of xenobiotics via the surface microlayer. Lipophilic organics accumulate there because of solubility, surface tension, and specific gravity considerations. This accumulation may also be critical to eggs and larvae of species that frequent this layer [Hardy et al. 1987]. The surface microlayer will be discussed further during this conference.

Suspended and Dissolved Organic Matter

Naturally occurring organic matter, e.g., humic acids, may sorb lipophilic xenobiotics. McCarthy [1983] reported that dissolved and colloidal organic matter reduced the bioavailability of PAHs to daphnia in laboratory experiments. The effect may be physical or chemical. The molecular size of the complex may limit bio-uptake, as mentioned previously. The chemical nature, e.g., the polarity, may also affect the bioaccumulation potential of the bound pollutant.

Carter and Suffet [1982] reported that DDT may sorb to dissolved humic materials and thus remain in the water column for extended periods of time. Parameters such as pH may affect the residence time of naturally occurring organic matter and, in turn, affect that of associated pollutants. Sorption of lipophilic xenobiotics to organics and particulates has been reported to have varying effects on persistence [Leslie et al. 1987]. Important routes of degradation include photochemical and microbial pathways.

Food

Organic pollutants may be available to biota via consumption of contaminated prey items or plant material. Ingestion of contaminated particulates was discussed previously. Biomagnification has been identified as a significant factor for the transmission of some persistent xenobiotics. For example, food has been observed to serve as the major vehicle for uptake of Kepone in blue crabs. Exposure of these crustaceans to Kepone via water, during laboratory experiments, resulted in minimal body burdens. However, when contaminated food (in the form of either James River or laboratory-exposed oysters) was provided, significant concentrations of Kepone were accumulated [Schimmel et al. 1979]. Obviously, differing feeding strategies will put certain organisms at greater risk, both because of the items selected, and because of the efficiency of uptake. The efficiency of transfer of lipophilic xenobiotics from the tissues of prey to consumers will be, as from the organic reservoirs of particulates, much less than that from water. Transfer of pollutants to humans via consumption of contaminated seafood is also of considerable concern.

SUMMARY OF BIOAVAILABILITY AND ASSESSMENT TECHNIQUES

A variety of techniques have been utilized to assess bioavailability. Laboratory procedures are appealing since variables may be more easily controlled. Bioaccumulation studies, using tissue burdens of parent compounds, suffer since biota are often able to metabolize xenobiotics which normally go undetected by conventional procedures. Radiotracers have the advantage that total xenobiotic material present in the organism may be determined; however, they are expensive, have limited availability, and require specialized handling. Bioaccumulation and toxicity studies often use environmentally unrealistic exposure scenarios, e.g., solvent carriers and filtered seawater, to obtain "reasonable results." Application of organic pollutants to sediments and immediate use in experiments ignores the effects of contact time on pollutant/sediment binding. The shortcomings of octanol/water determinations have been discussed. Adsorption isotherm determinations provide valuable information, although the observation that organic components of particulates are the major constituent into which lipophilic pollutants partition must be considered. Nonetheless, these techniques provide information critical to our understanding of the general processes controlling bioavailability.

Organic pollutants are readily accumulated by aquatic organisms. Many compounds exhibit appre-

ciable toxicity. Of particular concern are compounds of low solubility. These compounds may be bioaccumulated or biomagnified to high concentrations. They sorb to particulates, which may be ingested or settle to the bottom. Sediment-associated contaminants exhibit reduced bioavailability, but detrimental effects have been documented. Little information regarding the properties of the sediment/water interface is available, considering its importance with respect to pollutant fate and bioavailability. Differing feeding strategies and habitat selection may result in toxics being more available to some organisms than to others. Because the Bay is shallow, bioturbation, weather events, construction, boat traffic, and dredging may render sediment-adsorbed organics more available via re-working or resuspension.

The Chesapeake Bay represents a particularly complex theater for the investigation of the bioavailability question. Salinity regimes range from fresh to saline. These conditions may have measurable effects on the residence time of organic species and the physiology of aquatic organisms. Content of suspended particulates and content of colloidal and dissolved organics also differ drastically within the Bay. Tidal and riverine flows affect the disposition of toxics in both the water column and sediments. Turbidity, dissolved oxygen content, and temperature all influence the degradation rates of xenobiotics. These complexities will manifest themselves when laboratory data concerning bioavailability are applied to actual field situations.

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