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July 1995

U.S. Army Corps
of Engineers
Waterways Experiment
Station

Long-Term Effects of Dredging Operations Program

Evaluation of Field-Generated Accumulation Factors for Predicting the Bioaccumulation Potential of Sediment-Associated PAH Compounds

by Victor A. McFarland

LONG-TERM EFFECTS
OF DREDGING OPERATIONS PROGRAM

Prepared for Headquarters, U.S. Army Corps of Engineers

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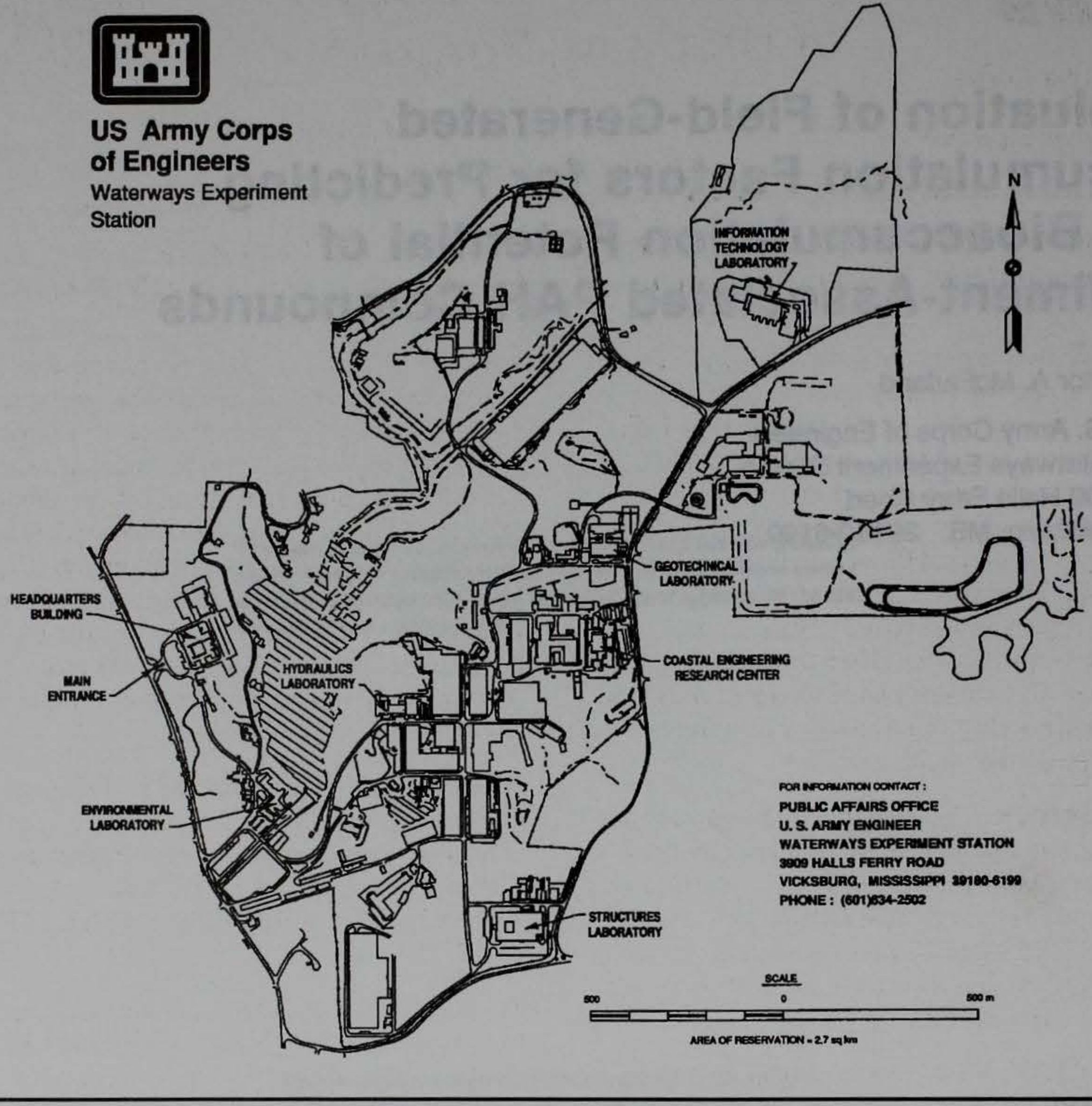

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LONG-TERM EFFECTS OF DREDGING OPERATIONS PROGRAM

Long-Term Effects of Dredging Operations Report Summary

Evaluation of Field-Generated Accumulation Factors for Predicting the Bioaccumulation Potential of Sediment-Associated PAH Compounds (TR D-95-2)

ISSUE: Before dredged sediments can be disposed in open water they must first be evaluated for potential adverse ecological effects including bioaccumulation of toxic chemicals. A widely used screening test for this purpose is based on sediment chemistry and organism lipid content. The test (Theoretical Bioaccumulation Potential, TBP) applies a universal accumulation factor, AF, to estimate expected body burdens in sediment-exposed organisms for all neutral organic chemicals, and all combinations of sediment and biota. This practice may be excessively conservative and not as predictive as assumed.

RESEARCH: This research was undertaken to test the hypothesis that AFs empirically determined for biota residing in PAH-contaminated sediments in their natural environment can be used in TBP calculations to predict bioaccumulation more accurately than is presently done using a universal AF.

SUMMARY: Field-collected benthic invertebrates and sediments from the New York Bight Apex were used to generate AFs for priority pollutant PAH compounds. The AFs were then used in TBP calculations to predict bioaccumulation potential of San Francisco Bay sediments. Correspondence between predicted and measured concentrations increased as the level of sediment contamination increased. In all cases, the field AF predictions were more accurate than predictions made using the universal AF. It was concluded that the use of field-generated AFs in TBP estimations markedly improves the predictive capability of the test for PAH compounds, and the same may be true of other bioaccumulating organic chemicals.

AVAILABILITY OF REPORT: The report is available on Interlibrary Loan Service from the U.S. Army Engineer Waterways Experiment Station (WES) Library, 3909 Halls Ferry Road, Vicksburg, MS 39180-6199; telephone (601) 634-2355.

To purchase a copy, call the National Technical Information Service (NTIS) at (703) 487-4780. For help in identifying a title for sale, call (703) 487-4780. NTIS report numbers may also be requested from the WES librarians.

About the Author: Dr. Victor A. McFarland is a research biologist in the WES Environmental Laboratory. For further information about the Long-term Effects of Dredging Operations Program, contact Mr. Thomas R. Patin, Program Manager, at (601) 634-3444.

July 1995

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LIST OF ABBREVIATIONS AND SYMBOLS

Abbreviation or symbol	Definition
Anth	anthracene
Acn	acenaphthene
Acnthy	acenaphthylene
AF	accumulation factor
Ar_2	argon gas
AU	below detection limit in all samples
B[a]A	benz[a]anthracene
B[a]P	benzo[a]pyrene
B[b+k]F	benzo[b+k]fluoranthene
B[ghi]P	benzo[g,h,i]perylene
BMSL	Battelle, Marine Science Laboratory
BSAF	biota/sediment accumulation factor
C	concentration
°C	degrees centigrade
C_b^{ℓ}	lipid-normalized concentration
Chry	chrysene
C_s^{oc}	organic carbon-normalized concentration
C_s	concentration in sediment
C_{ss}	concentration at steady state
C_t	concentration in tissue
C_w	concentration in water
cm	centimeter
D[ah]A	dibenz[a,h]anthracene
DCM	dichloromethane
DDT	dichloro-diphenyl-trichloroethane
DL	detection limit
f	fugacity
f_{ss}	fraction of steady state
F'	folded *F* test
FATES	Flow-through Aquatic Toxicology Exposure System
Fla	fluoranthene
Flu	fluorene
ft	foot
g	gram
gal	gallon
GC/MS	gas chromatography/mass spectrometry
H	Henry's Law constant

Abbreviation or symbol	Definition
ΔH	change in enthalpy
Hr	hour
HCl	hydrochloric acid
I[cd]P	indeno[1,2,3-cd]pyrene
J-value	quantitated value below detection limit
k_1	uptake rate constant
k_2	elimination rate constant
K_b	bioconcentration factor
Kg	kilogram
K_{oc}	sediment organic carbon/water partition coefficient
K_{ow}	octanol/water partition coefficient
LC_{50}	lethal concentration to 50 percent of population
L	liter
Lip	lipid
LSD	Fisher's Least Significant Difference Procedure
m	meter
m^2	square meter
MFO	mixed function oxidase
μL	microliter
μg	microgram
μm	micron
mg	milligram
min	minute
mL	milliliter
mm	millimeter
m.w.	molecular weight
n	number
N_2	nitrogen gas
NC	not calculated
Naph	naphthalene
ng	nanogram
P	oil/water partition coefficient
P (statistics)	probability
PAH	polynuclear aromatic hydrocarbon
‰	parts-per-thousand
PCB	polychlorinated biphenyl
PCDF	polychlorinated dibenzofurans
PCDD	polychlorinated dibenzo-*p*-dioxins
%ile	percentile

Abbreviation or symbol	Definition
PE	polyethylene
pf	preference factor
Phen	phenanthrene
Φ	fugacity coefficient
ppm	parts-per-million
Pyr	pyrene
RPD	relative percent difference
S	water solubility
SE	standard error of the mean
SF Bay	San Francisco Bay
SNK	Student-Neuman-Keul's Procedure
SOP	standard operating procedure
sp	species
SS	stainless steel
TBP	Theoretical Bioaccumulation Potential
TLm	median tolerance limit
TOC	total organic carbon
t	time
t	Student's *t*
tPAH	total of 15 PAH compounds
t_{ss}	time to steady state
TSS	total suspended solids
U	below detection limit in at least one sample
UPS	uninterruptable power supply
USACE	U.S. Army Corps of Engineers
USEPA	U.S. Environmental Protection Agency
WES	Waterways Experiment Station
WES/ALG	WES Analytical Laboratory Group
$\bar{x}$	mean
X^2	chi square

PREFACE

The work reported herein was conducted by the U.S. Army Engineer Waterways Experiment Station (WES) for the Headquarters, U.S. Army Corps of Engineers (HQUSACE), as part of the Long-term Effects of Dredging Operations Program (LEDO), Work Unit 31772. The LEDO Program is managed through the Environmental Effects of Dredging Programs, Dr. Robert M. Engler, Manager. LEDO Program Manager is Mr. Thomas R. Patin.

This report was prepared by Dr. Victor A. McFarland, Environmental Laboratory (EL), WES, in partial fulfillment of the requirements for a Ph.D. from Northeast Louisiana University (NLU), Monroe, LA. The author gratefully acknowledges the support provided by numerous individuals. Especially helpful were the WES Aquatic Contaminants Team members and the NLU advisory committee, including Dr. Paul W. Ferguson (Major Advisor) and Drs. Michael Crider, Pankaj Desai, and Benny Blaylock.

The work was conducted under the supervision of Dr. Bobby L. Folsom, Chief, Fate and Effects Branch, Mr. Donald L. Robey, Chief, Environmental Processes and Effects Division, and Dr. John W. Keeley, Director, EL.

At the time of publication of this report, Director of WES was Dr. Robert W. Whalin. Commander was COL Bruce K. Howard, EN.

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INTRODUCTION

Persistent organic chemicals released into the environment eventually become enriched in the sediments of waterways where they can be continually recycled through generations of aquatic biota. Chemicals such as the polychlorinated biphenyls (PCBs), polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDDs/PCDFs), chlorinated pesticides and polynuclear aromatic hydrocarbons (PAHs) are toxic to both aquatic organisms and their avian and terrestrial predators. Human consumers of contaminated fish and wildlife are also at risk. Decisions regarding remediation of contaminated waterways or disposal of dredged material for the maintenance of harbors and ship channels require assessing the ecological impacts of contaminants in sediments.

Both the Clean Water Act (Section 404 of PL 92-500) and the Marine Protection, Research, and Sanctuaries Act ("Ocean Dumping Act," Section 103 of PL 92-532) require determination of the potential for bioaccumulation of sediment-associated toxic chemicals using methods agreed upon by the U.S. Environmental Protection Agency (USEPA) and the U.S. Army Corps of Engineers (USACE), acting jointly. The procedures to be used for testing sediments are described in the Implementation Manual for Section 103 (the "Green Book") and in the draft Implementation Manual for Section 404 (the "Inland Manual") (U.S. Environmental

Protection Agency/U.S. Army Corps of Engineers 1991, 1994). These manuals share the same tiered approach and differ primarily in the specific conditions of testing appropriate to freshwater, estuarine and saltwater sediments and organisms.

Bioaccumulation testing according to the implementation manuals is a lengthy and expensive process. Organisms are exposed to bedded sediments for a period of 28 days, after which their tissues are analyzed for chemical contaminants. The 28-day standard exposure interval was adopted with the recognition that higher molecular weight neutral organic chemicals (log $K_{ow} \geq 4.7$) are not expected to reach steady-state bioaccumulation in that length of time (Connell 1990). However, for most bioaccumulating chemicals, 28-day exposures will result in a proportion of steady state sufficient to demonstrate the bioavailability, or lack of it, of chemicals associated with sediments. Tissue residues measured in test sediment organisms at the end of the exposure period are compared with residues in organisms exposed to an uncontaminated reference sediment. A decision against open water disposal of the dredged material may be made when the test sediment shows a greater bioaccumulation potential than does the reference. A decision for open water disposal will be influenced by low bioaccumulation potential as evidenced by comparability with the reference sediment. At least five replicate exposures of three

species of organisms to each test and reference sediment is recommended.

Chemical analyses of the tissues of organisms are costly, and when the presence of analytes such as PCDDs and PCDFs is suspected, the analytical costs can exceed $2,000/sample (Steve Calver, USACE District, Savannah; personal communication). Clearly, less costly alternatives to full-scale bioaccumulation testing are highly desirable.

BIOACCUMULATION POTENTIAL

A screening method for estimating the bioaccumulation potential of neutral organic chemicals recommended in both the Green Book and the Inland Manual is termed the "Theoretical Bioaccumulation Potential" (TBP) calculation. A TBP calculation uses sediment chemistry data to estimate the body burden of a neutral organic chemical in the tissues of an organism exposed to contaminated sediment if it were possible to achieve true equilibrium with the source of exposure. The magnitude of bioaccumulation of nonpolar organic compounds that could result by exposing aquatic organisms to the sediment is indicated (U.S. Environmental Protection Agency/U.S. Army Corps of Engineers 1991). TBP estimates are now commonly used in Tier II of dredged sediment evaluations to identify those sediments that are either sufficiently "clean" or sufficiently "contaminated" that they can be eliminated from further bioaccumulation testing at higher tiers. If the accuracy of TBP estimates

could be improved and extended to include organic compounds that may be metabolically degraded, it would become possible to eliminate much of the actual bioaccumulation testing currently being done.

The purpose of this dissertation is to:

- Describe the theoretical basis for TBP estimations,
- Calculate AFs for PAH compounds based on concentrations in sediments and biota collected in the field,
- Measure PAH bioaccumulation in organisms exposed in the laboratory to PAH-contaminated sediments,
- Estimate TBP for the contaminated sediments used in the laboratory exposures two ways:
  (1) Using the Green Book universal AF,
  (2) Using the field-generated AFs for specific PAHs,
- Compare estimations made using the two methods with concentrations of PAH compounds bioaccumulated in the laboratory exposures,
- Evaluate the relative predictive capability of TBP estimates made using the Green Book as opposed to a modification of the procedure using field data,
- Consider the sources of variability in environmental assessments that may affect the quality of predictions made using the model.

THEORY

THERMODYNAMICS

TBP is based in equilibrium partitioning theory. It was conceived as a thermodynamic model in which corrective terms that consider kinetic influences are to be added based on an iterative process of estimating and then comparing measured and predicted results.

The essence of equilibrium partitioning as applied to exposure of aquatic organisms to sediment-associated organic chemicals is that the relationship between sediment and organism can be idealized as a closed system in which distribution of an uncharged chemical occurs as a passive process, and the concentration of the chemical in each compartment of each phase of the system at equilibrium is dependent on the chemical activity of the phase (Karickhoff 1981, 1985).

Activity is defined as the ratio of the fugacity (escaping tendency) of a chemical at the state of interest with its fugacity at a chosen reference state (Prausnitz 1969). At equilibrium, fugacities, f_i, of a chemical are equal in all sorptive and solution phases (α, β, γ, etc.) of the system. The ambient fugacity is the product of chemical concentration, C_i, in each phase and a fugacity coefficient, Φ_i (Karickhoff 1981, 1985). That is,

$$f_\alpha = f_\beta = \ldots = f_\omega \qquad (1)$$

and for each phase

$$f_i = \Phi_i C_i. \quad (2)$$

If the reference states are chosen to be the same, then the equilibrium concentrations of chemical in any two phases of the system are described by a single constant that is a ratio of the fugacity coefficients of the chemical in the two phases. Because

$$f_\alpha = f_\beta = \Phi_\alpha C_\alpha = \Phi_\beta C_\beta \quad (3)$$

then

$$C_\alpha / C_\beta = \Phi_\beta / \Phi_\alpha = K_{\alpha\beta}. \quad (4)$$

In theory, it is possible to define a constant relationship for the equilibrium distribution of any chemical between any two phases. In practice, chemicals that interact coulombically or electrostatically in environmental matrices are nonlinear in their distributions between phases due to kinetic processes (Hamaker and Thompson 1972, Förstner 1990). Partitioning phenomena of charged or polar compounds are characterized by strong specific interactions between the sorbate/solute molecule and sorbent or solvent and involve increased enthalpies of sorption or solvation (ΔH) (Hassett et al. 1981, Karickhoff 1981, Prausnitz 1969). Although thermodynamics can give some indication of what to expect, the phase-distributions of metals, organometallic compounds, and speciating charged organics in heterogenous systems are difficult to predict

(Buffle et al. 1990, Rapin et al. 1986, Tessier et al. 1984).

However, the partitioning of neutral organic chemicals from water to organic or mineral phases is a relatively passive process involving predominantly van der Waals' interactions governing the association between sorbate and sorbent and are not accompanied by increased ΔH of sorption. Rather, sorptive associations between neutral organic chemicals partitioning from water, and non-aqueous phases such as mineral surfaces, sediment organic carbon, or organism lipids to which they partition, are thought to be entropy-driven by destruction of the highly energetic quasi-crystalline lattice structuring of water that occurs around uncharged molecules in aqueous solution (Horne 1978). Consequently, virtually all the chemical activity in an aqueous/organic system is accounted for by the energetic structuring of water, and the strength of sorption of organic solutes/sorbates is, then, more related to the size of the "hole" they make in water than to differences in their ability to form bonds with other neutral entities--a process termed "hydrophobic sorption" (Gustafson and Paleos 1971), or "hydrophobic bonding" (Hamaker and Thompson 1972). Horne (1978) described this behavior of water vividly:

> "The response of the water molecules to an intruder...is to 'join hands,' surround it, if possible exclude it, and at least try to minimize its volume, thereby minimizing the perturbation and masking its presence. This cooperative effort, not from attraction

> to one another but out of distaste for the alien, is not a feeble process. The so-called hydrophobic bond is so strong that it can dominate what we ordinarily consider as strong coulombic interactions."

This antipathy of water for the "alien", in the case of an uncharged molecule, motivates such a molecule to accept the first safe haven that becomes available. A safe haven being any surface or solvent that permits at least a partial reduction of contact with water. The lipids of aquatic biota, the organic carbon of humic origin in sediments, and to a lesser extent, the mineral surfaces themselves represent such safe havens for neutral organic molecules.

HYPOTHESIS

PARTITION COEFFICIENTS

In the period 1899-1901 H. Meyer and E. Overton showed that the narcotic activity of a homologous series of organic alcohols was correlated with their oil/water partition coefficients (P) (Hansch 1979). Since that time relative hydrophobicities (or lipophilicities) of biologically active organic compounds have been expressed using coefficients that relate their solubilities in a binary system consisting of immiscible solvents. The system most commonly used for this purpose consists of n-octanol and water, and the coefficient is usually expressed as log K_{ow}.

Examples of constants typically used in calculations of chemical environmental fate are octanol/water partition coefficients (K_{ow}), water solubilities (S), bioconcentration factors (K_b), Henry's Law constants (H), and soil or sediment organic carbon/water partition coefficients (K_{oc}). In each of these, water is the second phase. Using these coefficients, evaluative models can be constructed that calculate the concentrations in air, water, sediment, and biota that would be expected to result following the release of a known mass of an organic chemical into the environment (Mackay 1989, Mackay and Diamond 1989, Mackay and Paterson 1979, 1981, 1982; Mackay et al. 1983, 1985; Thomann 1989, Thomann and DiToro 1983, Thomann and Connolly 1984).

Partitioning of neutral organic chemicals to the organic carbon of sediments was first described by Lambert (1967) as being analogous to the solvation of such chemicals by water-immiscible solvents. Briggs (1973) showed that the analogy is appropriate by correlating the adsorption of un-ionized urea herbicides on soil organic matter with their K_{ow}s. Since that time it has been repeatedly shown that at the low concentrations typical of environmental contamination, the partitioning behavior of most neutral chemicals to water from soil or sediment, or from water to sediment or biota, varies linearly with concentration and can be described by constants unique to the individual solutes (Chiou 1985, Chiou et al. 1977; Gossett et al. 1983; Gschwend and Wu 1985, Hassett et al. 1980, 1981; Karickhoff 1981, Karickhoff et al. 1978; Mackay 1982, Veith et al. 1980).

Although the coefficients describing partitioning between water and non-aqueous phases for neutral organic compounds may differ by orders of magnitude, the partitioning to other organic phases is close to unity. This is illustrated by the solubilities of several organic compounds in organic solvents and in water shown in TABLE I. The four organic solutes listed are highly soluble in the organic solvents, ranging from 0.11 g mL^{-1} for DDT in olive oil or peanut oil, to 0.78 g mL^{-1} in toluene or benzene.

TABLE I

SOLUBILITIES OF ORGANIC SOLUTES IN ORGANIC SOLVENTS OR IN WATER[a], g/mL.

Organic Solute	Molecular Weight	Solvent				
		Toluene or Benzene	Olive Oil or Peanut Oil	$CHCl_3$	CCl_4	Water
Naphthalene	128.16	0.29	0.13	0.5	0.5	8.80E-7
Phenanthrene	178.22	0.42			0.42	1.06E-9
Lindane	290.85	0.29		0.24		1.70E-8
DDT	354.50	0.78	0.11		0.45	3.10E-12

[a]Merck (1983), Verschueren (1983).

Conversely, the water solubilities are seven to twelve orders of magnitude less and bear an inverse relationship with molecular weight, an approximation of molecular volume. *Similia similibus solvuntur* is an ancient concept in chemistry, but it is highly relevant to understanding the behavior of organic chemicals in the environment and to modeling their distributions among environmental phases.

THE PREFERENCE FACTOR

Theory suggests, as do the data of Table 1, that neutral organic solutes, in general, will have very similar solubilities in virtually any non-polar organic solvent. For neutral organic chemicals, a single constant will represent the partitioning between two organic phases within a reasonably small range of true values (Karickhoff 1981). For the purpose of modeling bioaccumulation from contaminated sediments in the aquatic environment, the two organic phases of interest are sediment organic carbon and organism lipid. The value of the idealized organic carbon/lipid distribution coefficient was termed a "preference factor," pf, and calculated based on regression equations obtained for two free-energy relationships in laboratory studies (McFarland 1984). In one study the organic carbon-normalized sorption, K_{oc}, of a series of PAH compounds by sediment particles was related to the octanol/water partition coefficients, K_{ow}, of the PAHs (Karickhoff 1981).

$$\log K_{oc} = 0.989 \log K_{ow} - 0.346, \quad r^2 = 0.987 \qquad (5)$$

In the second study the lipid-normalized bioconcentration factors, K_b, of a series of chlorobenzenes were related to their K_{ow} (Könemann and van Leeuwen 1980).

$$\log K_b = 0.980 \log K_{ow} - 0.063, \quad r^2 = 0.982 \qquad (6)$$

Highly linear regression estimates were obtained in both cases, with slopes near unity. Since K_{ow} is a common term in both equations, the Y-axis differential at the geometric mean log K_{ow} can be taken as an approximation of the difference in activity between the two organic phases and expresses the "preference" of neutral organic chemicals for lipid as opposed to sediment organic carbon. The antilog of the Y-axis differential, pf, was found to be 1.73 (95% CI: 1.48, 2.04) for the combined data sets (McFarland 1984, McFarland and Clarke 1986).

THEORETICAL BIOACCUMULATION POTENTIAL

On the basis of these relationships, a simple screening application was proposed to identify those sediments being considered for dredging and open-water disposal that could produce unacceptable levels of PCBs, DDT or other persistent neutral organic chemicals in exposed aquatic organisms. The "thermodynamically-defined bioaccumulation potential," TBP, suggested by McFarland (1984) applied the idealized pf to organic carbon-normalized concentrations of neutral chemicals in sediments in order to estimate the

concentration that could be expected to result in the lipids of an exposed organism at equilibrium. The model was described as,

> "... a simplification which has as its object determination of the maximum tissue concentration (C_i) that could be accumulated in organism tissue given sufficient time for equilibration."

and,

> "Kinetic, steric, or other constraints to release of chemicals from sediment and to absorption and elimination by organisms are not considered. Therefore the 'potential' thermodynamically defined is not necessarily the steady-state tissue concentration that could result from actual exposure."

The application was developed further by McFarland and Clarke (1986, 1987); and when the revised Green Book was published in February, 1991, it contained a new section in which it described "Theoretical Bioaccumulation Potential (TBP) of Non-polar Organic Chemicals." The section was essentially as described by McFarland and Clarke (1987), but instead of the pf (1.73) derived in the earlier work, a value of 4 was assigned, and the equation expressed as:

$$TBP = 4(C_s/\%TOC)\%L \qquad (7)$$

(C_s = concentration in whole sediment, TOC = sediment total organic carbon, and L = total extractable lipids in an organism of interest).

The higher value was termed an "accumulation factor," AF, and was chosen based on the results of field studies measuring PCB concentrations in infaunal bivalve mollusks

and polychaetes collected from contaminated sediments (Rubinstein et al. 1987).

ACCUMULATION FACTORS

The empirical relationship between lipid-normalized neutral organic chemical concentrations in biota and organic carbon-normalized concentrations in sediment is now most commonly referred to as an accumulation factor, AF (or biota-sediment accumulation factor, BSAF, and is expressed as:

$$AF = C_b^{\ell}/C_s^{oc} \quad (8)$$

in which C_b^{ℓ} is the tissue concentration normalized to percent or decimal fraction lipid, and C_s^{oc} is the concentration of the chemical in the sediments in the same units as C_b^{ℓ}, similarly normalized to percent or decimal fraction TOC in the sediment.

Empirical determinations have produced a range of AF values measured using different chemicals, organisms, sediments, and lengths of exposure. However, it is neither pf nor empirically derived AFs that are used in TBP calculations as recommended in the Green Book and Inland Manual. Rather, a universal AF = 4 based on early field studies is recommended for all applications. The universal AF has no ability to discriminate among chemicals, organisms, types of exposure, sediment characteristics, or any of the other possible sources of variability.

Consequently, TBP possesses only the crudest screening ability, and can only discriminate between sediments that are either virtually pristine, or grossly contaminated. The recommendation by the Green Book writers to use the AF = 4 suggested by PCB-based field studies rather than the pf calculated from laboratory studies was in keeping with the original concept of TBP which viewed the simple model as a first approximation. The model was to be recursively corrected by a process of estimating and comparing the result with the results of testing. Unfortunately, the next step has not yet been taken in the process, and the regulatory practice appears frozen with all TBP assessments being conducted assuming a four-fold difference.

Unquestionably, the current regulatory application of TBP is protective. However, an excessively protective screening test has little value; many sediments that fail the screen will subsequently be shown to result in negligible contaminant bioaccumulation when tests are done. Screening tests that are predictive as opposed to being merely protective have much greater value, in that they can prevent the unnecessary expenditure of limited resources.

Organisms exposed to chemicals in their natural environment provide the ultimate model for bioaccumulation potential in that they integrate all the opposing and augmenting influences on chemical release from sediments, uptake and elimination, and resultant body burden.

Therefore, this dissertation tested the hypothesis that AFs measured for specific chemicals in field-collected benthic organisms can be used in TBP calculations to more accurately predict bioaccumulation potential than can the Green Book universal AF = 4.

MATERIALS AND METHODS

APPROACH

Data of two studies (McFarland et al. [1994b,d]) performed at the U.S. Army Engineer Waterways Experiment Station (USAE WES) were used to generate and test AFs. A field study at a site on the continental shelf of the New York Bight Apex provided data that enabled calculation of AF values for 15 priority pollutant PAH compounds. In a concurrent laboratory study, tissue concentrations of the same PAH compounds were measured in bivalve mollusks after 28-day exposures to sediments from the San Francisco Bay (SF Bay) system. Exposures of bivalves in the laboratory were accomplished using the Flow-through Aquatic Toxicology Exposure System (FATES) of the WES Environmental Laboratory. Conditions of exposure were varied and involved both bedded and suspended sediments, and sediments having low and high degrees of PAH contamination.

TBP calculations were made using PAH concentrations in the SF Bay sediments to which clams or mussels were exposed. The estimates resulting from the Green Book TBP equation were compared with estimates using AFs for specific PAH compounds obtained from the data of the New York Bight Apex field study and with measured concentrations in the tissues of organisms exposed to the SF Bay sediments in the FATES for 28 days.

FIELD STUDY

Sediments and organisms were collected using a 0.1 m^2 Smith-McIntyre grab sampler at a location in the New York Bight Apex surrounding 40°20'29" North, 73°52'20" West. The collection period was 20-24 August, 1991.

Sediment samples

Sediment samples were sorted in plastic trays prepared by acid washing (10-percent HCl), hexane rinsing, and rinsing with site water prior to use. A 0.5 L aliquot of sediment from each grab sample was taken, and the composited samples were homogenized, large organisms were removed by hand, and five 1 L replicate samples were placed in acid-cleaned, hexane-rinsed glass jars with teflon lids. Jars were filled with no head space remaining. Four such composited samples were retained under refrigeration (4°C) for later analysis. At the end of the expedition, the sediment samples were shipped in coolers via overnight air freight to WES for physical and chemical characterization.

Biological samples

After sub-sampling for sediment analysis, the remainder of each grab sample was sorted in the field to collect organisms. Inorganic material was washed through 0.5 mm mesh sorting screens with collection site seawater to separate the organisms from the sediment. No single grab contained sufficient organisms of any taxonomic group to be treated as

a sample for chemical analysis. Therefore, organisms were collected and pooled by taxa in plastic sorting trays until a tissue mass sufficient for chemical analysis was judged to have been obtained. Species recommended in the Green Book were given priority for collection, but other species were also collected when found in sufficient tissue mass for chemical analysis.

Collected organisms were maintained alive in fresh collection site seawater until the end of each work day. Pooled organisms of a taxonomic group were then frozen in plastic Whirl-Pac® bags for analysis. All mollusks were frozen in their shells with the exception of *Mercenaria sp.* which were shucked on board the vessel because of their large size. Samples were stored in shipboard freezers at < 0°C, until shipment by overnight air freight to WES.

Samples were maintained frozen (-20°C) at the WES Environmental Laboratory on arrival. Sediment samples were homogenized and sent to the WES Analytical Laboratory Group (WES/ALG) on 9 October 1991. Tissues for PAH analysis were sent to the Battelle, Marine Science Laboratory (BMSL), Sequim, Washington, on 18 March 1992.

LABORATORY STUDIES

Two species of saltwater bivalve mollusks and a flatfish species were exposed together in the FATES. The two sediment composites used were a known contaminated sediment collected at a turning basin in Oakland Inner

Harbor, CA, (designated "Hot"), and a sediment representative of material normally resuspended in Central San Francisco Bay by wind or storm action, Berkeley Flats Reference (designated "Reference"). The Hot sediment was taken from an area that had previously been shown by chemical analysis to have elevated concentrations of metals and organic contaminants and was toxic to organisms in bioassays (Word et al. 1988). The Reference sediment consisted of material skimmed from the sediment/water interface at shoal areas in the eastern reaches of the Central Bay.

Sediment samples

Sediments were collected by BMSL. Collection expeditions were scheduled to precede FATES exposure experiments by 1-2 weeks to minimize storage time before use of each sediment. The Hot sediment consisted of a composite of single cores from two turning basin stations. Samples at the Hot stations were collected using a 12-inch Vibratory Hammer Corer. Total volume of each composite was 55 gal. Cores were mixed in an epoxy-lined drum aboard the collection vessel and were stored at 4°C. The Reference sediment was collected in a shoal area (2-3 m deep) of the East Bay using a boat-towed benthic sled device adjusted to skim the top few centimeters of sediment. Sediments were collected at two stations (122°19′18" W by 37°52′50" N and

122°18'40" W by 37°15'59" N). The collected material was similarly composited in an epoxy-lined 55-gal drum and stored at 4°C.

Sediment composites were shipped to WES by refrigerated truck (Hot sediment) or overnight air freight (Reference sediment) where they were maintained under refrigeration until used. Hot and Reference sediment exposures were performed sequentially and were begun immediately following preparation of the sediments. Before initiation of organism exposures, each sediment composite was homogenized using a large hand-held electric mixer, and five replicate one-L samples were taken for chemical analysis. All chemical analyses of Hot and Reference sediments and organisms were performed by BMSL. Hot sediment samples were sent to BMSL on April 17, 1991, and Reference sediments were sent on July 11, 1991.

Following homogenization, a high density slurry was prepared by mixing 120 gal N_2-sparged artificial seawater with ≈ 35 gal test sediment in a 200-gal polyethylene (PE) tank using a high shear-speed disperser with 316 stainless steel (SS) impeller and shaft. The slurry was sieved (3-mm) to remove debris, and pumped to a 675-L SS cone-bottom tank where it was continuously recirculated and maintained under Ar_2 to prevent oxidation. The remainder of the homogenized

sediment (≈ 15 gal) was reserved for use as bedded sediment in the FATES.

Organisms

Two invertebrate species (*Macoma nasuta* and *Mytilus edulis*) and a fish species (*Citharichthys stigmaeus*) were used as the experimental organisms. The three species are indigenous to San Francisco Bay and are representative of organisms that are abundant include the major feeding types and associations with bedded and suspended sediments and are ecologically and/or commercially important in the Bay system. Sanddabs, *C. stigmaeus*, are flatfish and are representative of fishes that live in contact with benthic sediments but respire, move, and feed in the water column. The bent-nosed clam, *M. nasuta*, is an infaunal species that actively ingests and processes sediments. *Mytilus edulis*, the Bay or blue mussel, is a fouling organism that filters large volumes of water (>3 L hr^{-1}) and by so doing concentrates chemical contaminants found in the water column. All organisms were collected from uncontaminated waters north of SF Bay (Brezina and Associates, Dillon Beach, CA). The animals were shipped by air express to WES and acclimated to experimental conditions for several weeks before the beginning of each experiment.

Acclimation

The acclimation facility consisted of a 10 x 20 ft photoperiod- and temperature-controlled building separated

from the experimental facilities. Organisms were maintained in fiberglass tubs in artificial seawater at 30 ‰ salinity during the acclimation period. The water was aerated and changed regularly, and sick or dead animals were removed. Sanddabs were fed a dry flake fish food and the bivalves received a liquid invertebrate diet. Organisms generally gained weight during acclimation. Only healthy animals, as evidenced by appearance and feeding and avoidance behavior, were used in the experiments.

Three pooled replicate samples of each acclimated species were frozen in glass for lipid determination and trace chemical analysis at the start of exposures.

Exposure system

A large-capacity aquarium system (FATES) with continuous once-through replacement of water provided exposures of the three species to the sediments. Exposures were to bedded or to suspended sediment maintained at constant concentrations. The system consisted of 24 round-bottomed 75-L circular aquaria, each with its own recirculating pump and transmissometer probe (Figure 1). Clean water and test sediment slurry entered through an inflow port at the top of each aquarium. Flow-through water replacement was established by the pulsed addition of clean make-up water and the removal of an equal volume through an overflow port opposite the point of water entry. Water was mixed and recirculated by withdrawal through a screened

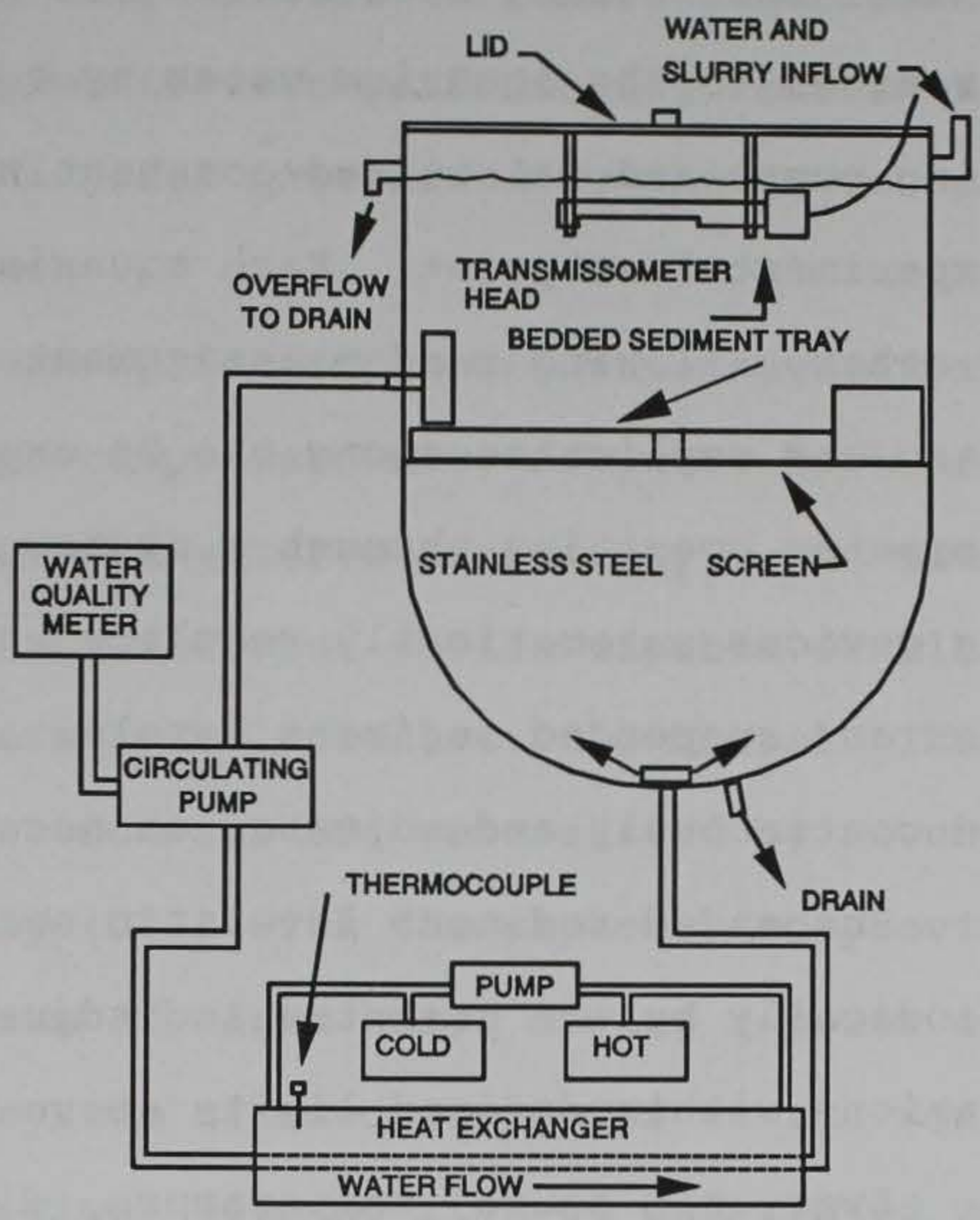


Figure 1. Schematic representation of one FATES aquarium.

suction port in the side of each aquarium, pumped through the closed-system heat exchanger, and then back into the bottom of the aquarium. All but the largest sediment particles were kept in suspension by the resulting bottom-to-top current flow. When configured in suspended sediment-only mode, a stainless-steel screen floor prevented fish from having contact with any particulate material that settled out of suspension.

Two heat exchangers, each serving 12 aquaria, removed heat introduced to the aquarium water by action of the recirculating pumps and maintained constant water temperature at the experimental setpoint. Each aquarium was independent of every other, allowing random assignment of controls, treatments, and replicates among the 24 experimental units.

A microcomputer operating through a system of sensors and switching devices automatically regulated temperature, water flow rate, and suspended sediment levels. Temperature was monitored continuously and adjusted as necessary by the computer. Suspended sediment levels in each aquarium were read periodically by the computer and adjusted to maintain concentrations within defined limits above and below the setpoint. Every six hours, temperature, dissolved oxygen, pH and conductivity data were reported for each aquarium. These data were stored on disc and automatically printed at set intervals, or on demand, allowing for continuous monitoring of the experimental conditions. Photoperiod and salinity were controlled externally.

All water used by the FATES was collected in a sump and then pumped through a particulate filter that removed a large fraction of the sediment particles from the water. The collected sediment was retained in a steel drum for disposal. The remaining sediment and water flowed into a settling basin and the particle-free effluent was then pumped through a series of bag filters, activated carbon

filters and clay filters to remove solubilized contaminants. Settled sediments were periodically removed from the sump for waste disposal determined by the degree of contamination.

An alarm system monitored water availability, electricity, compressed air, computer software errors, water temperature, and water conductivity. The alarm system contained an autodialer device capable of alerting investigators by telephone at all hours. The computer and data acquisition systems were served by an uninterruptable power supply (UPS) with the capacity to provide electricity for a minimum of eight hours during a power outage. The UPS and alarm system allowed the FATES to run continuously for extended periods with no risk of undetected major breakdowns.

Exposure conditions

The environmental conditions established for the SF Bay Hot and Reference sediment exposures were 30 ‰ salinity artificial seawater, 15°C water temperature, and 12 hr/12 hr day/night light cycle over 28 days of exposure. A flow-through water replacement mode was maintained with the addition of ≈ 350 mL seawater into each aquarium every two min. This rate of delivery produced > 99% replacement/8 hr. Sufficient organisms of each species were placed in each aquarium so that at least 50 grams of tissue were available for analysis at the end of the exposure. The approximate

numbers of organisms used in each aquarium for each experiment were 80 fish, 20 mussels, and 20 clams. Numbers of fish varied with average fish size, ranging 55 to 99 fish per aquarium.

At the end of the exposure periods organisms were exchanged to clear flowing water for 24 hrs for elimination of ingested or entrained sediment. After purging, clams and mussels were shucked, the shells discarded, and species from each aquarium were pooled separately in acid-washed and hexane-rinsed glass jars. All tissue samples were preserved for analysis by freezing.

System performance

Gravimetric total suspended solids (TSS) measurements (U.S. Environmental Protection Agency 1979) were performed three times weekly on each aquarium during setup and exposures as a manual check on the performance of the automated suspended sediment control system. Physical data (temperature, dissolved oxygen, and salinity) were also measured manually twice weekly and were compared to the computer-monitored values. Discrepancies between the manual data and the computer data were verified and equipment calibrations performed as necessary. Dead or injured organisms were removed and organisms were fed during daily visual inspections.

Water supply

A commercial marine aquarium sea salt (Instant Ocean™) was used to make up the experimental water. A stock brine solution of 90 ‰ sea salts was made using a high shear-speed mixer and stored in a 2000 gal PE tank. Water was pumped upon activation of a low-limit float switch to a second 2000 gal tank and diluted 2:1 with aged tap water, producing the required experimental salinity. Both storage tanks were internally recirculated to maintain salts in solution. The artificial seawater was pumped on demand through a sand filter to a small head tank and dispensed by a computer-timed valving system to the FATES aquaria.

Suspended sediment

A transmissometer probe mounted beneath the plexiglas lid of each aquarium read underwater light transmissance at a sub-surface depth of 8 cm at 15 min intervals. This measurement was compared to preset levels in a computer program, and metered amounts of stock slurry were added to maintain suspended sediment concentrations near the setpoint. Setpoints were established before the start of exposures by calibrating transmissometer output against simultaneous gravimetric measurement of TSS in each aquarium.

Experimental design

In each of the experiments, FATES aquaria were randomly assigned treatments and replications:

(1) Laboratory (negative) controls, three replicates.

(2) Positive controls, three replicates.

(3) Bedded test sediment with clear water flow-through, six replicates.

(4) Flow-through suspension of test sediment fines at approximately 10 mg L^{-1} without bedded test sediment, six replicates.

(5) Flow-through suspension of test sediment fines at approximately 50 mg L^{-1} without bedded test sediment, six replicates.

Control treatments

Two variations of laboratory control were used because of the length of the experimental runs and the necessity of continuing the experiments over an inactive period in the winter and spring. Negative laboratory controls consisting of clear culture water over washed sand or gravel substrate were included for two purposes: first, as a check on the overall environmental quality of the system, and second, to detect any contaminant uptake by organisms caused by the experimental facilities themselves. Positive controls were included in these experiments to provide a measure of the constancy of exposure among experiments. The positive controls consisted of clear culture water over inert

substrate, as with the negative controls, but also included continuous metering of solubilized chemicals into the aquaria to maintain constant exposure concentrations. The chemicals included single representatives of major classes of bioaccumulating sediment contaminants.

Experimental treatments

The 18 remaining aquaria were configured to provide simultaneous exposure to the three species in an environment containing either bedded test sediment or suspended test sediment, but not both. The mussels were kept in coarse-mesh net containers near the top of an aquarium. Clams were allowed to bury themselves in mesh-covered sediment trays. Fish were free to swim in the aquaria and to settle on the sediment surface in the bedded sediment treatments or the perforated SS aquarium floor in suspended sediment treatments.

Bedded sediment treatment

Six replicates were prepared. A 46-cm diameter plexiglass tray, 5 cm deep in the open sediment area, and 10 cm deep in the screened clam area was filled to a depth of ≈ 2 cm (open area) and 6 cm (screened area) with test sediment, and was placed on the SS mesh floor of an aquarium. The clams were allowed to burrow in the deeper sediment section and a high density PE netting (5 mm mesh) was fastened over them to prevent predation by the fish. Mussels were suspended in the upper water column in a PE net

bag. The fish were added last, and their access to the bedded sediment was unrestricted.

Suspended sediment, 10 mg L^{-1} treatment

A 10 cm-deep plexiglass compartment identical to the clam area of the sediment trays in the bedded sediment treatment was used in the six replicates of the low suspension treatment. The compartment was filled to a depth of ≈ 5 cm with uncontaminated sediments that had been collected with the clams. The compartment was covered with PE netting and placed on the SS mesh floor of the aquarium. Mussels were suspended in the water column in PE mesh bags as in the bedded sediment treatment. Fish were free in the aquaria but denied access to any bedded sediment. The clams burrowed in the uncontaminated sediment but fed on the surface where some settling of the suspended test sediment occurred. The only contact the fish or mussels had with the test sediment was via the water column where suspended sediment levels were maintained at ≈ 10 mg L^{-1}.

Suspended sediment, 50 mg L^{-1} treatment

The same aquarium configuration was used in the six replicates of the higher suspension treatment, and the concentration of suspended sediment in the water column was maintained at ≈ 50 mg L^{-1}.

Negative control

Three aquaria were configured as for the suspended sediment exposures. Clear water only was used, and no test sediment (bedded or suspended) was available to the three species.

Positive control

Three bioaccumulating chemicals (DDT, phenanthrene, and cadmium) in seawater solution were continuously added to the three positive control aquaria using a chemical metering pump. The calculated dosage was sublethal, but sufficient to bioaccumulate. Configuration of the aquaria was the same as for the negative controls.

CHEMICAL ANALYSIS

Polynuclear aromatic hydrocarbons in sediments were extracted with dichloromethane (DCM) (BMSL) or 1:1 acetone/hexane (WES/ALG) according to EPA Method 3540 (U.S. Environmental Protection Agency 1986) followed by clean-up on alumina and copper (BMSL) or silica gel (WES/ALG following Warner 1976). Tissues were extracted in DCM using a roller under ambient conditions following SOP MSL-M-42. Samples were cleaned up using silica/alumina (5% deactivated) chromatography (Krahn et al. 1988). Extracts were analyzed for individual PAH compounds following EPA Method 8270 (U.S. Environmental Protection Agency 1986) by gas chromatography/mass spectrometry (GC/MS) using a Hewlett Packard HP 5890 GC (BMSL) or an HP 5880 GC (WES/ALG) both

equipped with an HP 5970 MS detector. Oven temperature was programmed to increase at a rate of 6°C min^{-1} from 35°C to the final temperature of 325°C. The carrier gas was He at an approximately 25 cm sec^{-1} flow-rate. The column used was a J&W DB-5 30 m, 0.25 mm i.d., and 0.25 μm film thickness (BMSL), or a Hewlett Packard Ultra 2 column 25 m, 0.32 mm i.d., and 0.52 μm film thickness (WES/ALG).

Recoveries ranged 40 to 120%, relative precisions were ± 30%, and detection limits were 10 ng g^{-1} except in sediment samples analyzed by WES/ALG, which ranged 670-1000 ng g^{-1}. Low recoveries were the result of smaller than optimal sample sizes obtained in several of the field-collected taxa. All tissue concentration data are reported on a wet weight basis. All sediment concentration data are reported on a dry weight basis.

Lipids were determined on 100 μL aliquots of the DCM extract (removed prior to clean-up steps) prepared for the PAH analyses. The residue remaining after drying was weighed in a tared pan on a Cahn C31 electronic microbalance. Lipids were reported as a percentage of the sample wet weight.

Total organic carbon (TOC) in the sediment samples was determined using a DC-80 total carbon analyzer equipped with a sludge and sediment sampler accessory.

DATA REDUCTION

Initial data analysis and reduction for statistical analysis and graphing was conducted using Quatro® Pro Version 4.0 (Borland International, Inc. 1992). Graphics were performed on Freelance Plus™ (Lotus Development Corporation 1986) and SigmaPlot™ Version 5.0 (Jandel Scientific 1992). The report was prepared using WordPerfect Version 5.1 (WordPerfect© Corporation 1990).

STATISTICAL ANALYSIS

Concentration data were summarized using means and standard errors. Standard errors of field-derived AFs were calculated using bootstrap estimation (Manly 1991). Twenty-eight day bioaccumulation data and TBP estimations using field-derived AFs or using Green Book (AF = 4) calculations were compared using Fisher's Least Significant Difference (LSD) procedure, Student-Newman-Keul's Procedure, and Tukey's *W* procedure. Prior to making comparisons, the normality assumption was tested using Shapiro-Wilk's test (Shapiro and Wilk 1965), and the equality of variances assumption was tested using Levene's test (Snedecor and Cochran 1989) or *F'* (folded *F*) test (SAS Institute, Inc. 1988a,b). When parametric testing hypothesis assumptions were not met, the data were converted to rankits and the same comparison methods were used (LSD, SNK, Tukey). Analysis of variance employed PROC GLM (General Linear Measures Procedure, SAS Institute, Inc. 1988a,b).

In the evaluation of comparability of exposures from bedded and suspended sediment, 28-day AFs were calculated using means of three to six replicates. Standard errors of the AFs were estimated using bootstrap resampling (Manly 1991) with 1024 resamples per calculation. AF comparisons between bedded sediment and suspended sediment groups for each organism were conducted using *t*-tests when the residuals were normally distributed, and Kruskal-Wallis tests when they were not. Observations below detection limit were excluded from 28-day AF calculations, and 28-day AFs were determined only for analytes having at least three replicate observations above detection limit. All statistical analyses and bootstrap estimations of standard error of the AFs were performed using SAS® (SAS Institute Inc. 1988a,b).

RESULTS

CONCENTRATION DATA

The conventions used in treating the reported concentration data for calculation of descriptive statistics and statistical analysis, and for calculating AFs, were the same in both the field and the laboratory studies. Many of the data were reported as less than the detection limit (DL). When observations less than DL were reported as actual quantitated values (i.e., "J values"), and they were within a factor of ten of the DL, those data were used in the same way as data above the DL. All other values reported as < DL and all J values < 1/10th of the detection limit were set equal to DL/10 for inclusion in the statistical analyses. Outliers occurred frequently and were not deleted unless they were an obvious error.

Data were considered acceptable for statistical analysis when surrogate percent recovery was within two standard deviations of the mean percent recovery for that surrogate, or when laboratory-specified quality control criteria were not exceeded.

A number of samples were split into duplicates (sometimes triplicates) as a laboratory quality control check. When the relative percent difference (RPD) between laboratory duplicates was within the acceptable quality control criteria range, the mean of the duplicate values was used in the statistical analyses. If the RPD was outside

the acceptable quality control criteria range, then the mean of the duplicates was used if both duplicate values fell within the range of values for other replicates of the same treatment. Otherwise, the duplicate value was used that was within or closest to the range of values for other replicates of the same treatment.

When data were flagged by the analytical laboratory because the contaminant analyte was present in a blank, those data were considered biased and were not included in the statistical analyses.

Complete data sets, including all quality control/quality assurance data, can be found in McFarland et al. [1994b,d].

FIELD STUDY

During the three-day sampling excursion, 343 grab samples were taken at the New York Bight Apex station. The appearance of the sediment samples was generally similar, with changes in sediment color, type, and associated fauna being gradual over the sampling area. The coarser sediments yielded relatively depauperate faunal assemblages, compared to the finer grained material. Only fine-grained, dark colored material yielded *Nucula sp.* *Nucula sp.* were collected from only six grab samples, but when found were collected by the hundreds.

Few crustaceans were found at the site, with approximately half being small isopods, and these were

insufficient for residue analysis. Several species of amphipods, primarily *Corophium* and *Ampelisca sp.*, were collected, but also were not present in sufficient tissue mass for analysis.

The largest taxonomic group collected at the site in terms of abundance and tissue mass was the nemertean, *Cerebratulus lacteus*. These annelids are entirely carnivorous (Barnes 1974) and do not ingest or process sediments directly. However, because of their abundance at the site, and their importance in the benthic food web, they were included in the chemical analyses. The primary prey of nemerteans are small polychaete infaunal species, and polychaete to nemertean trophic transfer of contaminants was expected to occur.

The organisms collected and the wet weights of the pooled samples are given in TABLE II. Taxa present in sufficient tissue mass for analysis were: *Nucula sp.*, *Mercenaria sp.* (probably *M. mercenaria*), *Cerebratulus lacteus*, and *Nephtys sp.* (probably *N. incisa* and/or *N. picta*). Remaining organisms were further pooled into broader taxonomic groups to provide sufficient tissue for analysis in each group. These were: Mollusca, Polychaeta, and Lumbrineridae. Numbers of replicate samples for analysis ranged, depending on total tissue mass, from two replicates for *Mercenaria sp.* to five each for Lumbrineridae and Polychaeta.

Mean concentrations of individual priority PAH compounds in the eight New York Bight Apex species or taxonomic groups are given in TABLE III. Concentrations ranged from 0.14 ng g^{-1} indeno[1,2,3-cd]pyrene in *Mercenaria sp*. to 113 ng g^{-1} fluoranthene in *Nucula sp*. The concentration of benzo[b+k]fluoranthene was 163 ng g^{-1} in *Nucula sp*., but represented the total of two isomers. In some cases, concentrations of a PAH were consistent across organisms. For example, naphthalene ranged about 20-30 ng g^{-1} for all organisms. However, in most cases the concentrations of individual PAH compounds ranged widely across species and related more to feeding type and relationship with the sediment than to lipoidicity of the organism. *Mercenaria sp*. and *Nucula sp*. were both near 1.0 percent total DCM-extractable lipid (0.88 and 1.15 percent, respectively). However, the filter-feeding clam, *Mercenaria sp*. was typically among the lowest in body burden of PAH compounds with tissue concentrations of less than 10 ng g^{-1} in nearly all cases. *Nucula sp*., which is an infaunal polychaete and a sediment-ingesting organism, generally had body burdens of the same PAH compounds in the range 20-100+ ng g^{-1}.

Concentrations of PAH compounds analyzed in the four composites of the collection site sediments are reported in TABLE IV. Accumulation factors (TABLE V) were calculated

TABLE II

BENTHIC ORGANISMS AND WEIGHTS (g) OF POOLED SAMPLES COLLECTED AT THE NEW YORK BIGHT APEX SITE.

Polychaetes		Crustaceans	
Nephtys sp. (incisa or picta)[a]	119	Isopods	5
unknown worm parts	105	Assorted	8
Pherusa sp. (affinis)	24		
Lumbrineridae	393	Mollusks[b]	
Glycera sp.	48	*Nucula sp.*	94
Diopatra cuprea	12	unsorted mollusks (primarily *Nucula* and shell hash)	459
orbiniidae	21	*Astarte sp.*	198
opheliidae	2	*Nassarius trivittatus*	4
Sigalion arenicola	6	*Ensis correctus*	44
		Yoldia limatula	6
Nemerteans		*Spisula solidissima*	62
Cerebratulus lacteus	82	*Mercenaria sp. (mercenaria)*	60

[a]Species names in parentheses are tentative identifications.
[b]Mollusk weights include shells.

TABLE III

CONCENTRATIONS OF PAH COMPOUNDS AND LIPIDS IN ORGANISMS COLLECTED AT THE NEW YORK BIGHT APEX SITE. MEANS (STANDARD ERRORS)

Taxonomic Group	Concentration, ng g^{-1} wet weight			
	Naph[a]	Acn[b]	Acnthy[c]	Flu[d]
Nucula sp.	23.8 (4.33)	17.6 (9.95)U[f]	7.06 (0.43)	16.8 (9.54)U
Mercenaria sp.	34.6 (0.00)	0.60 (0.16)U	0.30 (0.00)U	0.54 (0.09)U
Mollusca	27.5 (3.56)	0.98 (0.23)U	0.51 (0.11)U	0.88 (0.12)U
Nephtys sp.	20.6 (1.08)	1.27 (0.23)U	0.71 (0.15)U	1.05 (0.23)U
C. lacteus	28.0 (2.68)	1.08 (0.18)U	0.61 (0.10)U	0.91 (0.15)U
Lumbrineridae	29.1 (1.97)	0.93 (0.11)U	0.52 (0.06)U	0.81 (0.09)U
Polychaeta	29.1 (1.97)	0.80 (0.16)U	26.2 (3.48)U	0.80 (0.17)U

[a]Naphthalene
[b]Acenapthene
[c]Acenaphthylene
[d]Fluorene
[f]U indicates below detection limit in at least one sample.

TABLE III (CONTINUED)

Taxonomic Group	Concentration, ng g^{-1} wet weight			
	Anth[a]	Pyr[b]	Fla[c]	Chry[d]
Nucula sp.	23.9 (11.9)	101 (36.0)	113 (55.8)	60.3 (20.5)
Mercenaria sp.	0.36 (0.06)U[e]	8.59 (0.20)	7.40 (0.19)	5.07 (0.17)
Mollusca	2.42 (1.76)U	31.6 (2.04)	19.1 (1.38)	19.5 (1.28)
Nephtys sp.	2.82 (2.54)U	8.85 (1.08)	4.06 (2.00)U	2.95 (1.61)U
C. lacteus	0.60 (0.10)U	3.98 (1.72)U	0.45 (0.08)	2.55 (1.04)
Lumbrineridae	0.54 (0.06)U	14.7 (1.10)	7.89 (0.63)	10.6 (0.74)
Polychaeta	1.04 (0.45)U	15.0 (0.28)	6.60 (0.06)	9.27 (0.20)

[a]Anthracene
[b]Pyrene
[c]Fluoranthene
[d]Chrysene
[e]U indicates below detection limit in at least one sample.

TABLE III (CONTINUED)

Taxonomic Group	Concentration, ng g^{-1} wet weight			
	B[a]P[a]	D[ah]A[b]	B[b+k]F[c]	B[ghi]P[d]
Nucula sp.	50.8 (17.7)	20.3 (6.96)	163 (55.0)	47.9 (16.5)
Mercenaria sp.	0.23 (0.04)U[e]	0.17 (0.03)U	5.76 (0.12)	2.75 (0.07)
Mollusca	15.6 (1.44)	3.18 (0.05)	32.7 (2.49)	13.4 (1.76)
Nephtys sp.	1.52 (1.24)U	0.30 (0.07)U	7.03 (2.39)U	2.47 (1.44)U
C. lacteus	1.25 (0.81)U	0.29 (0.06)U	12.0 (1.35)	2.45 (1.05)U
Lumbrineridae	12.5 (1.08)	3.48 (0.32)	32.2 (2.22)	9.49 (0.78)
Polychaeta	9.37 (0.28)	2.56 (0.54)	23.3 (2.09)	8.65 (0.18)

[a]Benzo[a]pyrene
[b]Dibenz[a,h]anthracene
[c]Benzo[b+k]fluoranthene
[d]Benzo[g,h,i]perylene
[e]U indicates below detection limit in at least on sample.

TABLE III (CONTINUED)

Taxonomic Group	Concentration, ng g^{-1} wet weight			
	Phen[a]	B[a]A[b]	I[cd]P[c]	tPAH[d]
Nucula sp.	90.5 (46.7)	52.8 (19.6)	43.8 (16.6)	822 (298)
Mercenaria sp.	8.18 (1.46)	4.10 (0.03)	0.14 (0.02)U	52.4 (32.1)
Mollusca	15.8 (2.92)	14.0 (0.66)	6.26 (0.71)	203 (17.7)
Nephtys sp.	6.42 (3.06)U[e]	0.43 (0.10)U	1.04 (0.48)U	53.2 (12.9)
C. lacteus	10.6 (0.79)	0.38 (0.07)U	0.80 (0.53)U	65.5 (3.94)
Lumbrineridae	11.5 (0.91)	8.80 (0.88)	7.89 (0.54)	151 (9.89)
Polychaeta	10.1 (1.11)	7.34 (0.59)	6.31 (0.26)	155 (6.91)

[a]Phenanthrene
[b]Benz[a]anthracene
[c]Indeno[1,2,3-cd]pyrene
[d]Total of 15 PAH compounds
[e]U indicates below detection limit in at least one sample.

TABLE III (CONCLUDED)

Taxonomic Group	Lipid, percent wet weight[a]
Nucula sp.	1.15 (0.14)
Mercenaria sp.	0.88 (0.03)
Mollusca	4.78 (0.34)
Nephtys sp.	3.52 (0.14)
C. lacteus	3.70 (0.92)
Lumbrineridae	1.43 (0.29)
Polychaeta	4.22 (0.31)

[a]Total dichloromethane-extractable lipids

TABLE IV

CONCENTRATIONS OF PAH COMPOUNDS IN SEDIMENTS COLLECTED AT THE NEW YORK BIGHT APEX SITE. MEANS (STANDARD ERRORS)

Compound	Concentration ng g^{-1} dry weight	
Naphthalene	42.5	(2.50) AU[a]
Acenaphthene	42.5	(2.50) AU
Acenaphthylene	42.5	(2.50) AU
Fluorene	42.5	(2.50) AU
Phenanthrene	34.0	(16.1)
Anthracene	42.5	(2.50) AU
Pyrene	20.0	(4.30)
Fluoranthene	40.0	(17.4) U[b]
Chrysene	24.5	(5.66) U
Benz[a]anthracene	25.8	(6.54) U
Benzo[a]pyrene	42.5	(2.50) AU
Dibenz[a,h]anthracene	42.5	(2.50) AU
Benzo[b+k]fluoranthene	86.8	(87) U
Benzo[g,h,i]perylene	42.5	(2.50) AU
Indeno[1,2,3-cd]pyrene	42.5	(2.50) AU
Total of 15 PAH	571	(79.3)

[a]AU indicates below detection limit in all samples
[b]U indicates below detection limit in at least one sample.

using concentration data of the field study. The sediment PAH concentrations of TABLE IV were normalized on organic carbon content in each of the four replicates. Concentrations of PAH compounds in the individual tissue samples (TABLE III) were normalized on lipid content of the sample. Seven taxonomic groups having two to five replicate analyses each were treated similarly, resulting in 24

TABLE V

POLYNUCLEAR AROMATIC HYDROCARBON ACCUMULATION FACTORS (AF) BASED ON CONCENTRATIONS IN SEVEN TAXA AND SEDIMENT COLLECTED AT THE NEW YORK BIGHT APEX SITE. MEANS[a] (STANDARD ERRORS[b]) BIAS[c]

Compound	AF	(SE)	Bias
Naphthalene	O.1031	(0.0357)	- 0.0059
Acenaphthene	0.0104	(0.0053)	- 0.0007
Acenaphthylene	0.0233	(0.0096)	- 0.0012
Fluorene	0.0027	(0.0008)	- 0.0001
Phenanthrene	0.0435	(0.0377)	- 0.0154
Anthracene	0.0085	(0.0047)	- 0.0005
Pyrene	0.1477	(0.0592)	- 0.0080
Fluoranthene	0.0415	(0.0262)	- 0.0060
Chrysene	0.0614	(0.0443)	- 0.0100
Benz[a]anthracene	0.0425	(0.0322)	- 0.0079
Benzo[a]pyrene	0.0343	(0.0166)	- 0.0020
Dibenz[a,h]anthracene	0.0108	(0.0056)	- 0.0007
Benzo[b+k]fluoranthene	0.0731	(0.0750)	- 0.0019
Benzo[g,h,i]perylene	0.0324	(0.0135)	- 0.0016
Indeno[1,2,3-cd]pyrene	0.0225	(0.0112)	- 0.0014
Total of 15 PAH	0.0415	(0.0434)	- 0.0019

[a]Pooled lipid-normalized data of seven taxa (24 replicates)/pooled organic carbon-normalized data of four sediments (four replicates)

[b]Bootstrap standard errors, 1024 replicates

[c]Difference between bootstrap mean and sample mean.

analyte/lipid replicates for each PAH compound. The AF for each PAH was calculated using the means of the four normalized sediment and 24 tissue replicates.

Because AFs are ratios of means with unequal *n* in the numerator and denominator terms, the conventional variance calculation procedure was not considered appropriate. A

non-standard method for variance estimation, the "bootstrap" considers that the true population is approximated by an infinite population in which each of the observed experimental values is equally likely. A large number of random samples from the population of experimental values, each of size *n*, is taken and the standard error is calculated from those data (Manly 1991). Additionally, the difference between the bootstrap mean and the sample mean is taken as an estimation of sampling bias. The bootstrap estimations are included in TABLE V, and are based on 1,024 random samplings of each normalized PAH compound data set.

LABORATORY STUDY

No system shut-downs occurred during the 28-day Hot or Reference sediment exposures. No major excursions from nominal settings were recorded for any of the system performance parameters. Temperature was maintained at 15 ± 0.75°C and dissolved oxygen remained high, greater than 7.5 mg L^{-1}. Salinity and pH remained stable over the course of both experiments. Total suspended solids concentrations were near the target concentrations of 10 and 50 mg L^{-1}, averaging ≈ 20 and 60 mg L^{-1}.

Of the 15 PAH analytes only four were less than DL in one or more of the Reference sediment samples (i.e., naphthalene, acenaphthene, acenaphthylene, and dibenz[a,h]anthracene). All analytes were present above DL

in all of the Hot sediment samples. Concentration data of the two sediments are shown in TABLE VI. Pyrene and fluoranthene were present in highest concentrations in both sediments, but were approximately 30-fold higher in the Hot than in the Reference sediment. With a few exceptions (e.g., acenaphthene, fluorene, dibenz[a,h]anthracene), concentrations of the 15 PAH analytes in the Reference and Hot sediments were approximately proportional, with Hot sediment concentrations on the order of 20 to 30-fold higher than Reference sediment concentrations (r^2 = 0.9764).

Organic carbon content of the Reference and Hot sediments was similar (means: 0.926 and 1.110 percent; standard errors: 0.007 and 0.085 percent, respectively).

Only tissue samples of the higher suspended sediment treatment (≈ 50 mg L^{-1}) of the Reference and Hot sediment experiments were analyzed. When the flatfish were analyzed, few PAH were present above DL in their tissues, and it was decided not to use the four fish treatment combinations for TBP comparisons. Consequently, the data set of the laboratory study uses only the bivalve data and consists of eight organism-sediment-exposure regime combinations. The treatment combinations are shown in TABLE VII.

The lipid content of the bivalves at the end of 28-day exposures in each treatment combination are given in TABLE VIII. Clams and mussels both lost lipid during the exposures, ranging from 2.54% lost by mussels in the bedded

TABLE VI

CONCENTRATIONS OF PAH COMPOUNDS IN SAN FRANCISCO EAST BAY (REFERENCE) AND OAKLAND HARBOR TURNING BASIN (HOT) SEDIMENTS USED IN 28-DAY EXPOSURES
MEANS (STANDARD ERRORS)

Compound	Concentration, ng g^{-1} dry weight			
	Reference Sediment		Hot Sediment	
Naphthalene	19.2	(4.91) U[a]	550	(38.0)
Acenaphthene	1.62	(0.023)U	1239	(111)
Acenaphthylene	5.29	(3.68) U	69.3	(10.8)
Fluorene	1.62	(0.026)U	534	(79.9)
Phenanthrene	111	(16.4)	5053	(459)
Anthracene	27.3	(4.02)	1766	(27.3)
Pyrene	252	(18.5)	7330	(651)
Fluoranthene	242	(17.9)	7122	(661)
Chrysene	106	(8.95)	3204	(327)
Benz[a]anthracene	116	(9.65)	2409	(259)
Benzo[a]pyrene	193	(16.7)	4306	(193)
Dibenz[a,h]anthracene	5.09	(3.48) U	434	(40.2)
Benzo[b+k]fluoranthene	223	(602)	7368	(14.5)
Benzo[g,h,i]perylene	128	(306)	3268	(15.0)
Indeno[1,2,3-cd]pyrene	127	(300)	3600	(11.7)
Total of 15 PAH	1558	NC[b]	48252	NC

[a]U indicates below detection limit in at least one sample
[b]Not calculated.

TABLE VII

TREATMENT COMBINATIONS OF THE 28-DAY LABORATORY STUDY

Number	Sediment	Organism	Exposure regime
1	Reference	Clam	Bedded sediment
2			Suspended sediment
3		Mussel	Bedded sediment
4			Suspended sediment
5	Hot	Clam	Bedded sediment
6			Suspended sediment
7		Mussel	Bedded sediment
8			Suspended sediment

Reference sediment exposure to 25.6% lost by clams in the suspended Reference sediment exposure. No relationships were discernible regarding type of exposure or organism and percentage of lipid lost.

Bioaccumulation

Day-28 tissue concentrations of the contaminants were divided by the day zero concentrations in each treatment and the logarithms of the ratios are plotted in Figures 2-9. Benzo[b]fluoranthene and benzo[k]fluoranthene were quantitated separately in tissue analyses of the San Francisco Bay study and are not combined in the Figures. Bars to the left of the zero line indicate decreases in body burden during the exposure period, and in some cases these were statistically significant ($P_{\alpha/2} \leq 0.025$). Mussels and

TABLE VIII

LIPID[a] CONTENT OF CLAMS (*M. NASUTA*) AND MUSSELS (*M. EDULIS*) EXPOSED TO SAN FRANCISCO EAST BAY (REFERENCE) AND OAKLAND HARBOR TURNING BASIN (HOT) SEDIMENTS IN 28-DAY EXPOSURES. MEANS (STANDARD ERRORS)

Sediment	Organism	Lipid, percent wet weight			
		Bedded sediment exposure		Suspended sediment exposure	
Reference	Clam	1.47	(0.212)	1.22	(0.111)
	Mussel	2.30	(0.111)	2.11	(0.101)
Hot	Clam	2.99	(0.606)	3.08	(0.493)
	Mussel	1.50	(0.108)	1.64	(0.073)

[a]Total dichloromethane-extractable lipids.

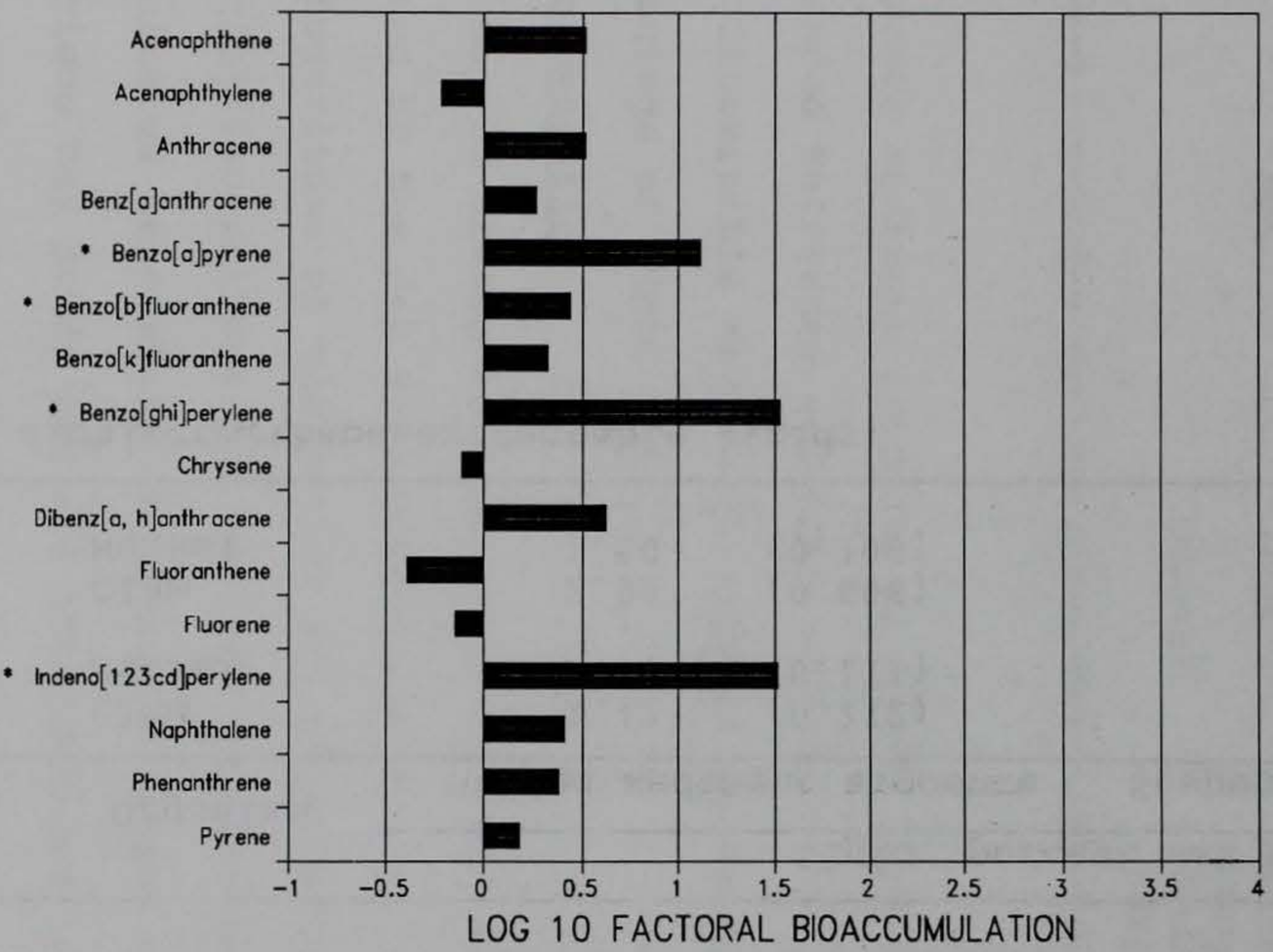


Figure 2. Bioaccumulation in clams, *M. nasuta*, exposed to bedded Reference sediment for 28 days. Asterisk indicates statistical significance, $P_{\alpha/2} \leq 0.025$

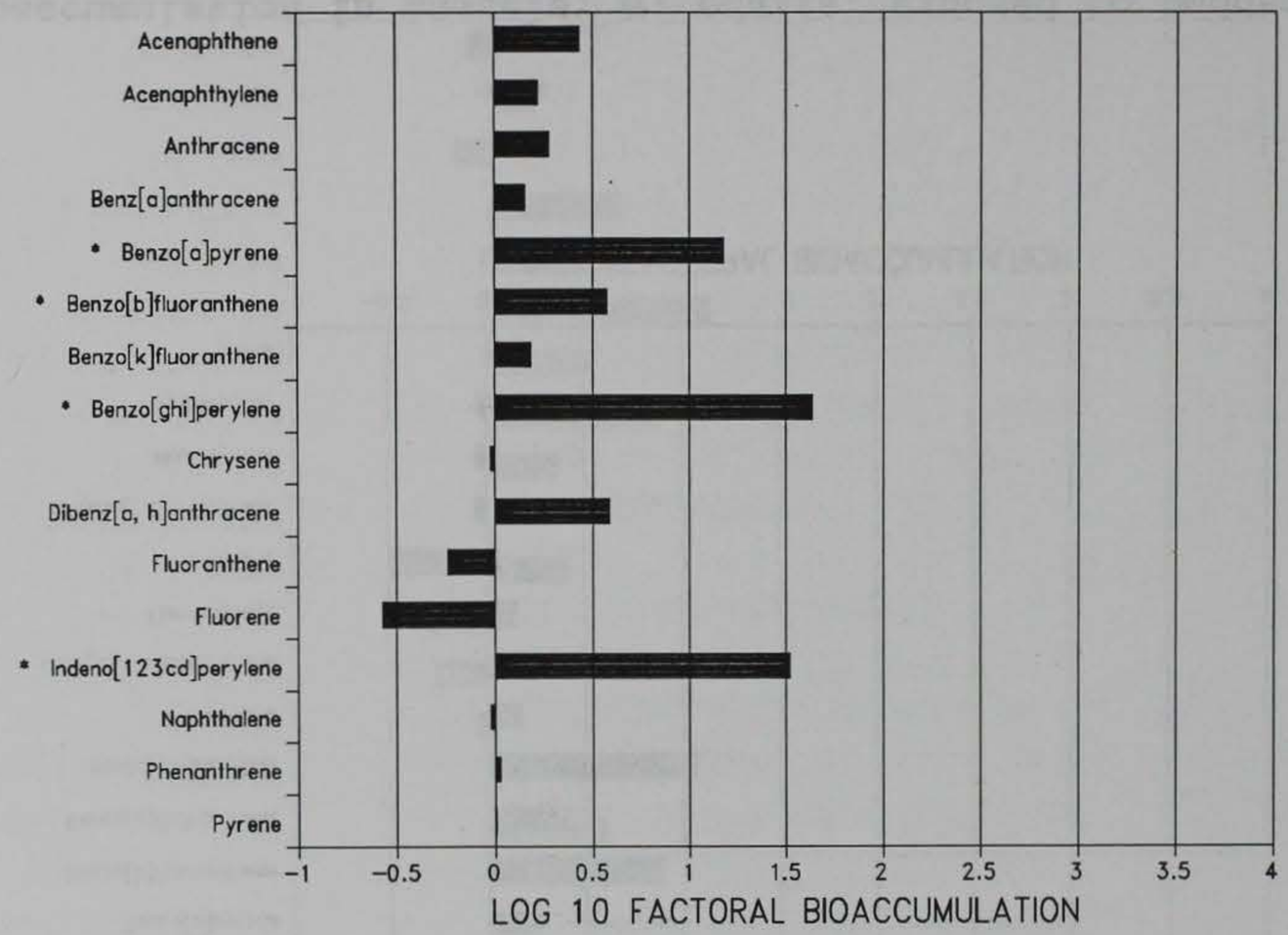


Figure 3. Bioaccumulation in clams, *M. nasuta*, exposed to suspended Reference sediment for 28 days. Asterisk indicates statistical significance, $P_{\alpha/2} \leq 0.025$.

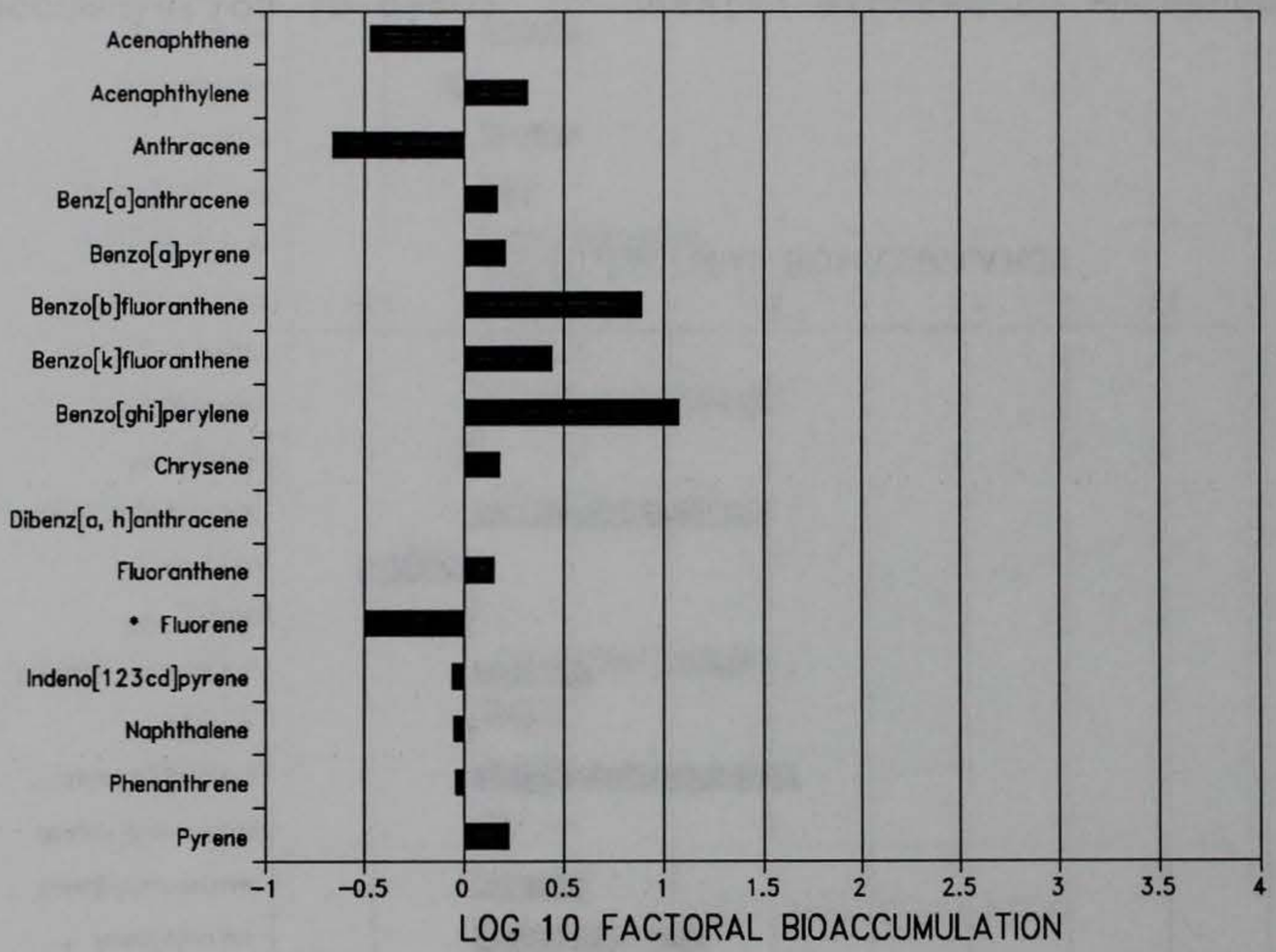


Figure 4. Bioaccumulation in mussels, *M. edulis*, exposed to bedded Reference sediment for 28 days. Asterisk indicates statistical significance, $P_{\alpha/2} \leq 0.025$.

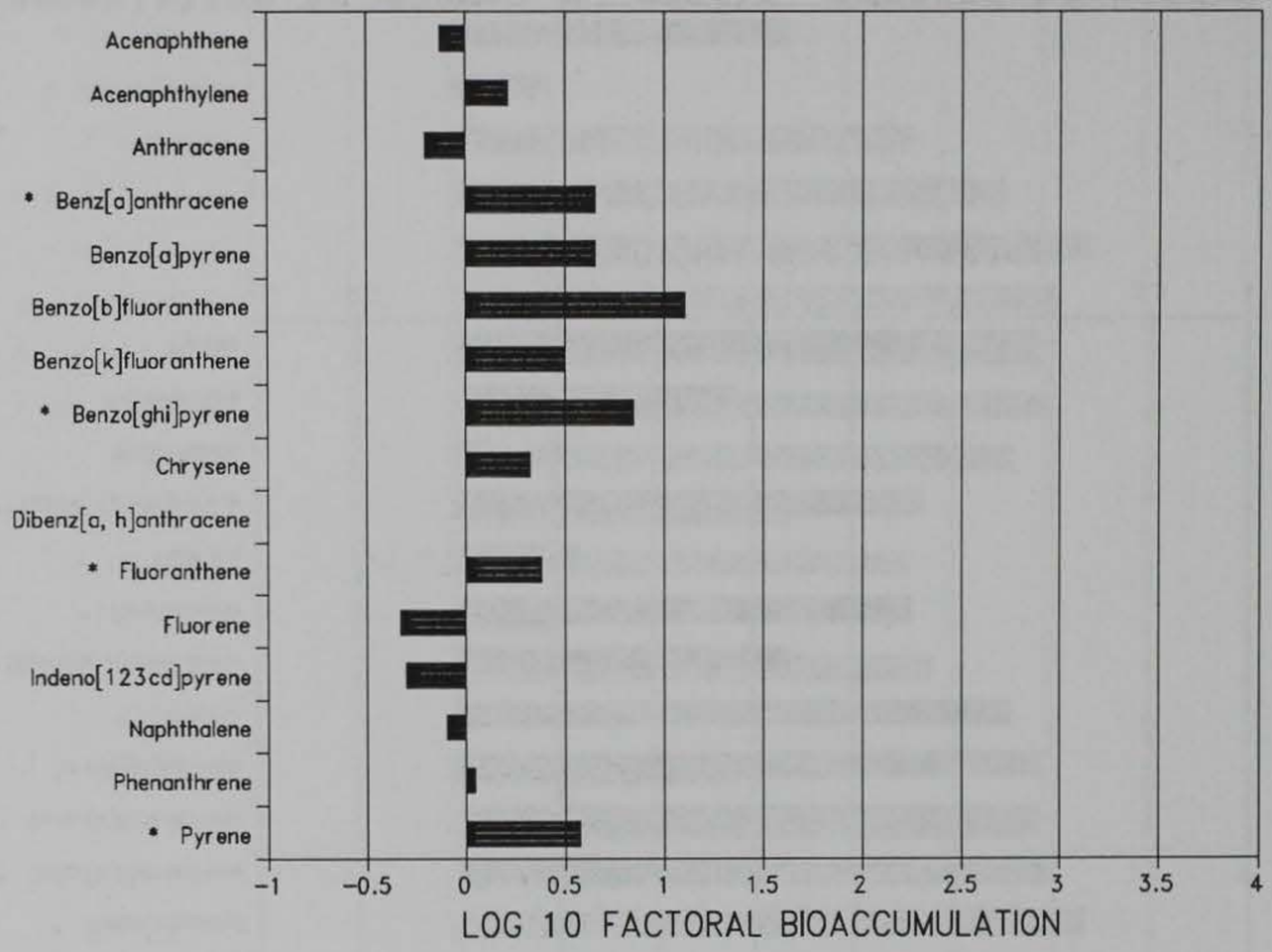


Figure 5. Bioaccumulation in mussels, *M. edulis*, exposed to suspended Reference sediment for 28 days. Asterisk indicates statistical significance, $P_{\alpha/2} \leq 0.025$.

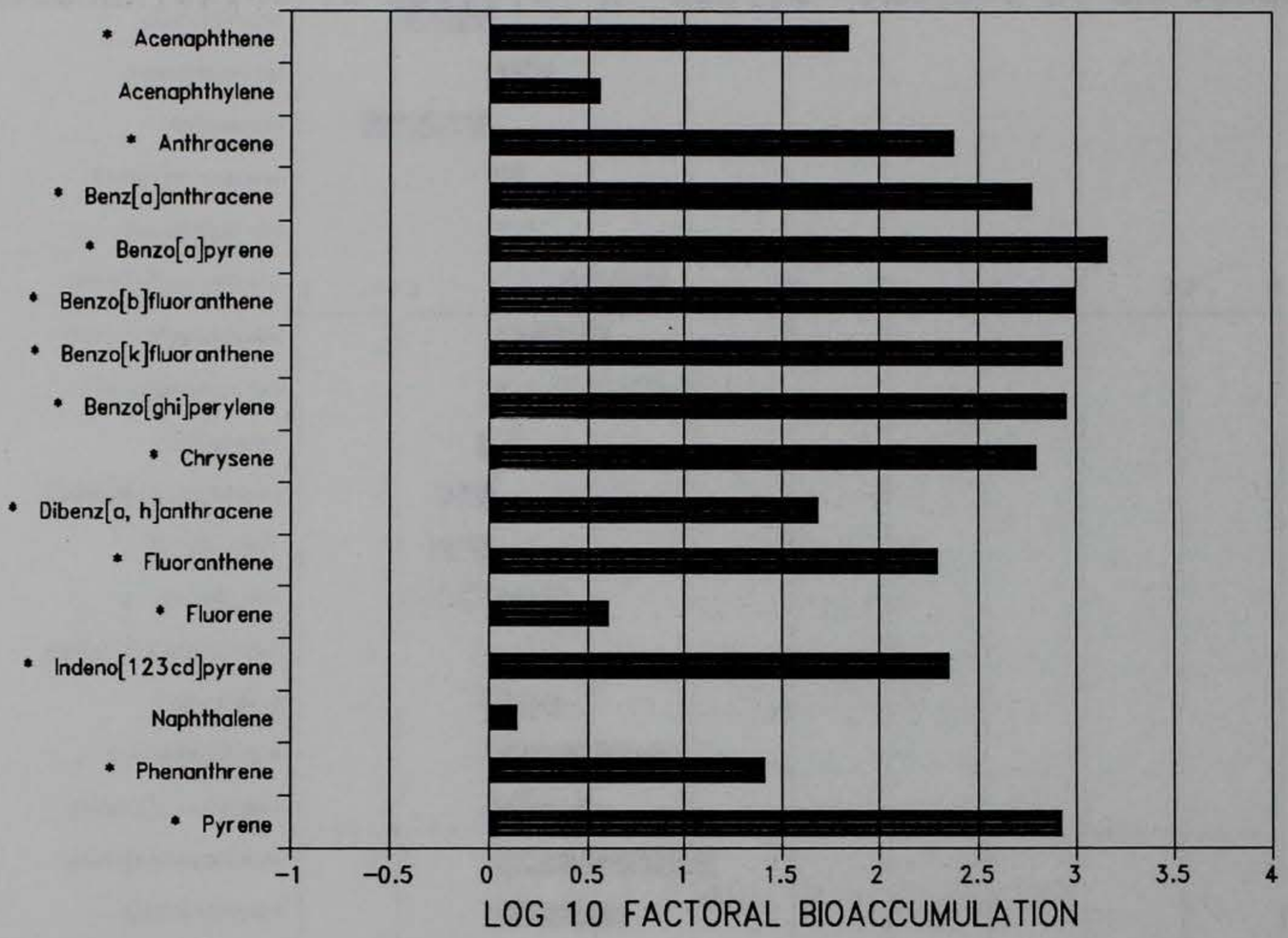


Figure 6. Bioaccumulation in clams, *M. edulis*, exposed to bedded Hot sediment for 28 days. Asterisk indicates statistical significance, $P_{\alpha/2} \leq 0.025$.

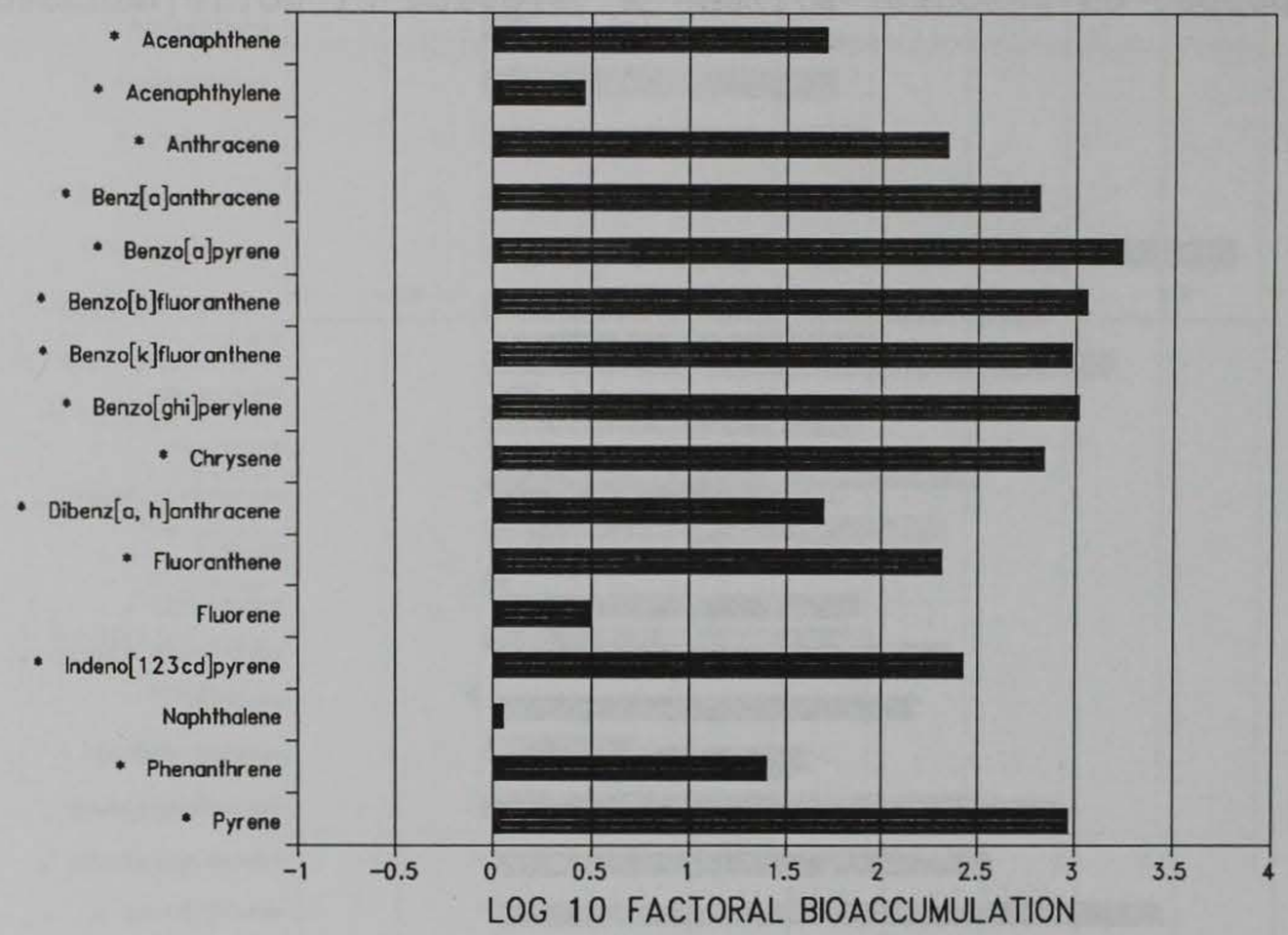


Figure 7. Bioaccumulation in clams, *M. nasuta*, exposed to suspended Hot sediment for 28 days. Asterisk indicates statistical significance, $P_{\alpha/2} \leq 0.025$.

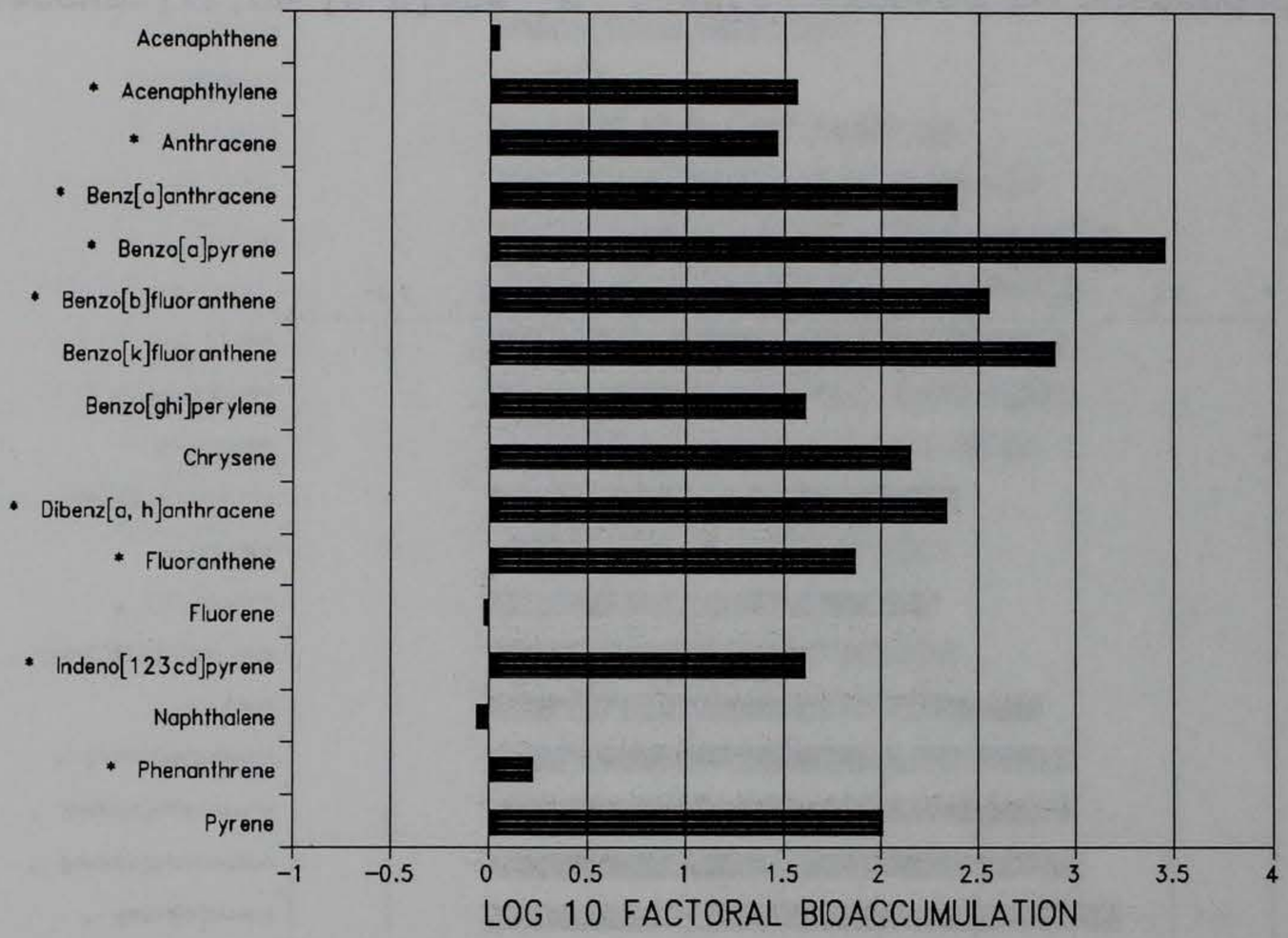


Figure 8. Bioaccumulation in mussels, *M. edulis*, exposed to bedded Hot sediment for 28 days. Asterisk indicates statistical significance, $P_{\alpha/2} \leq 0.025$.

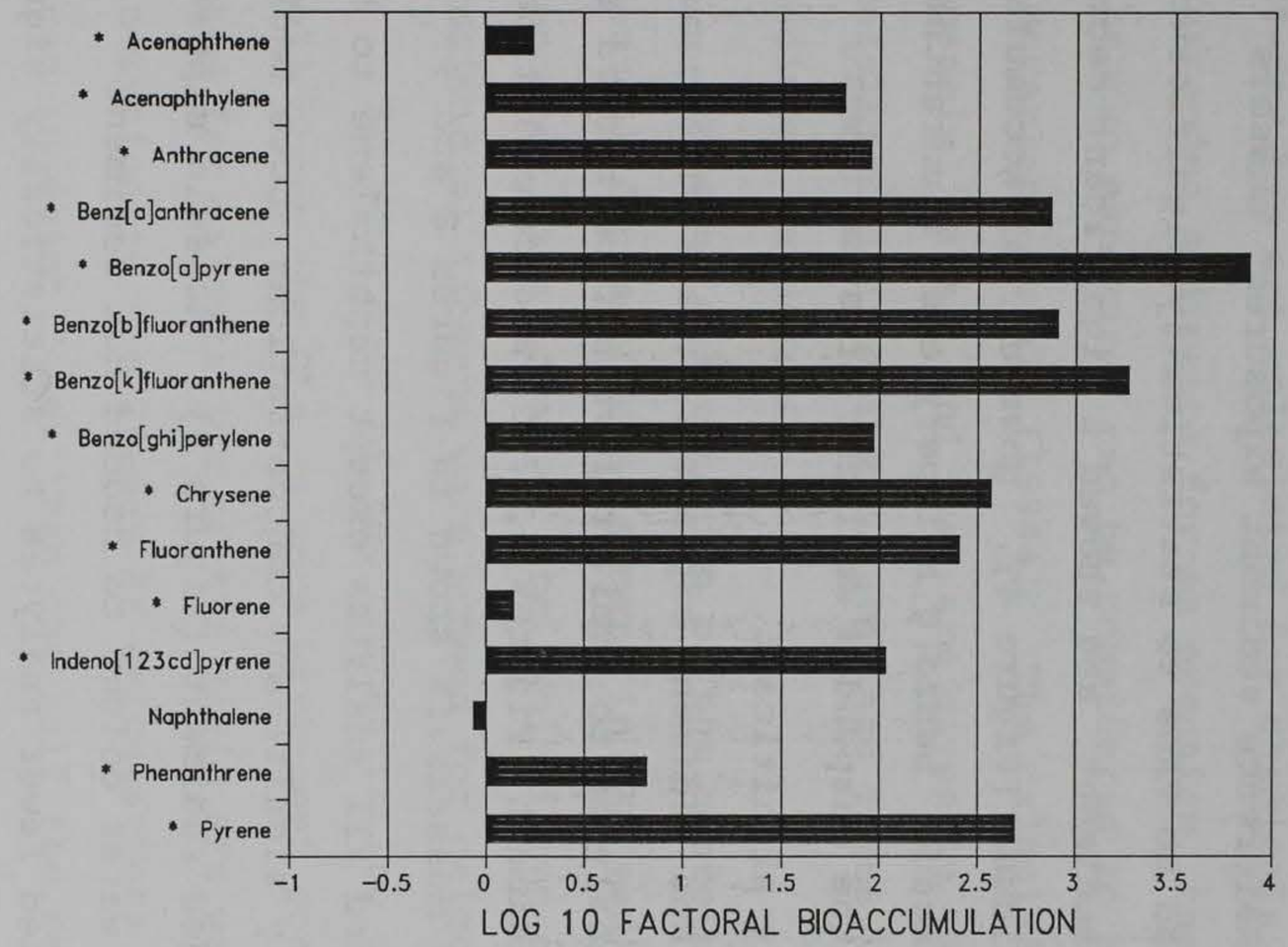


Figure 9. Bioaccumulation in mussels, *M. edulis*, exposed to suspended Hot sediment for 28 days. Asterisk indicates statistical significance, $P_{\alpha/2} \leq 0.025$.

clams generally showed similar patterns of uptake in the bedded and suspended sediment exposures.

Clams bioaccumulated four PAHs to significant levels from both bedded and suspended Reference sediment exposures: benzo[a]pyrene, benzo[b]fluoranthene, benzo[g,h,i]perylene, and indeno[1,2,3-cd]pyrene (Figures 2, 3).

In the Reference sediment exposures, mussels bioaccumulated no PAHs to statistically significant levels from bedded sediment, and showed a significant decrease in fluorene residues (Figure 4). However, bioaccumulation of benz[a]anthracene, benzo[g,h,i]perylene, fluoranthene, and pyrene from the suspended sediment treatment were statistically significant.

In the Hot sediment exposures, clams bioaccumulated nearly all analytes to similar levels from bedded and suspended sediment (Figures 6, 7). Bioaccumulation of PAH compounds by mussels is shown in Figures 8 and 9. Mussels bioaccumulated all analytes except naphthalene to higher and statistically significant concentrations above background from suspended sediment (Figure 9). Mussels exposed only through the water column to bedded Hot sediment bioaccumulated fewer analytes to statistically significant levels (9/15 PAH compounds as opposed to 14/15 in the suspended sediment).

28-Day accumulation factors

In order to compare relative bioavailability of PAH compounds to the bivalves from bedded as opposed to suspended sediments, 28-Day AFs were calculated using the mean concentrations of chemical, tissue lipid, and sediment TOC at the end of the 28-day exposure periods. If similar 28-day AFs are calculated for chemicals in bedded and in suspended sediments for an organism, the bioavailability in the two exposures can be considered to be similar. (McFarland et al. [1994a]. The calculated 28-day AFs for mussels, clams, and fish are given in TABLE IX for the Reference sediment exposures and TABLE X for the Hot sediment exposures.

Because quantitation of the benzo[b]- and benzo[k]fluoranthene isomers in the sediment samples was reported as a total of the two, tissue concentrations of the individual isomers were summed for calculation of the AF.

Because most of the concentration data were reported well above the DLs, a more stringent convention than was used for calculation of the Field AFs was adopted for data inclusion in the 28-day AF calculations. No treatments were used in which fewer than one-half the replicates were reported above the DL or as "J values." In the Reference sediment exposures, at least four of the six replicate sediment or tissue analyses of all organism/bedded sediment or organism/suspended sediment combinations resulted in

TABLE IX

TWENTY-EIGHT DAY PAH ACCUMULATION FACTORS FOR CLAMS AND MUSSELS EXPOSED TOGETHER TO EITHER BEDDED OR SUSPENDED REFERENCE SEDIMENT. MEANS (STANDARD ERRORS)

Compound	Mussel (*M. edulis*) 28-Day AF (SE)		Clam (*M. nasuta*) 28-Day AF (SE)	
	Bedded	Suspended	Bedded	Suspended
B[a]A[a]	NC[k]	NC	0.028 (0.0044)	0.026 (0.0031)
B[a]P[b]	NC	NC	0.015 (0.0023)	0.021 (0.0021)
B[b+k]F[c]	0.007 (0.0006)	0.010 (0.0007)	0.028 (0.0039)	0.040 (0.0038)
B[ghi]P[d]	NC	NC	0.022 (0.0048)	0.035 (0.0044)
Chry[e]	0.012 (0.0018)	0.023 (0.0051)	0.025 (0.0037)	0.038 (0.0047)
Flu[f]	0.010 (0.0014)	0.019 (0.0018)	0.023 (0.0033)	0.041 (0.0048)
I[cd]P[g]	NC	NC	0.021 (0.0030)	0.025 (0.0025)
Naph[h]	0.839 (0.1520)	0.827 (0.1305)	1.060 (0.2573)	0.475 (0.1277)
Phen[i]	0.090 (0.0152)	0.121 (0.0179)	0.114 (0.0292)	0.060 (0.0123)
Pyr[j]	0.012 (0.0021)	0.028 (0.0032)	0.022 (0.0035)	0.042 (0.0049)

[a]Benz[a]anthracene
[b]Benzo[a]pyrene
[c]Benzo[b+k]fluoranthene
[d]Benzo[g,h,i]perylene
[e]Chrysene
[f]Fluoranthene
[g]Indeno[1,2,3-cd]pyrene
[h]Naphtnalene
[i]Phenanthrene
[j]Pyrene
[k]Not calculated

TABLE X

TWENTY-EIGHT DAY PAH ACCUMULATION FACTORS FOR CLAMS AND MUSSELS EXPOSED TOGETHER TO EITHER BEDDED OR SUSPENDED HOT SEDIMENT. MEANS (STANDARD ERRORS)

Compound	Mussel 28-Day AF (SE)		Clam 28-Day AF (SE)	
	Bedded	Suspended	Bedded	Suspended
Acn[a]	0.003 (0.0004)	0.005 (0.0005)	0.016 (0.0028)	0.012 (0.0032)
Acnthy[b]	0.065 (0.0126)	0.109 (0.0193)	NC[n]	NC
Anth[c]	0.006 (0.0015)	0.016 (0.0040)	0.050 (0.0154)	0.044 (0.0139)
B[a]A[d]	0.064 (0.0096)	0.175 (0.0208)	0.070 (0.0122)	0.074 (0.0098)
B[a]P[e]	0.040 (0.0062)	0.097 (0.0084)	0.028 (0.0039)	0.032 (0.0044)
B[b+k]F[f]	0.056 (0.0079)	0.118 (0.0105)	0.032 (0.0043)	0.035 (0.0052)
B[g,h,i]P[g]	0.017 (0.0024)	0.036 (0.0032)	0.011 (0.0016)	0.013 (0.0020)
Chry[h]	0.084 (0.0117)	0.196 (0.0202)	0.073 (0.0124)	0.079 (0.0116)
Fla[i]	0.032 (0.0045)	0.102 (0.0118)	0.093 (0.0168)	0.095 (0.0134)
I[cd]P[j]	0.011 (0.0016)	0.026 (0.0027)	0.007 (0.0010)	0.009 (0.0013)
Naph[k]	0.079 (0.0079)	0.071 (0.0055)	0.044 (0.0099)	0.035 (0.0085)
Phen[l]	0.007 (0.0011)	0.018 (0.0025)	0.047 (0.0093)	0.044 (0.0090)
Pyr[m]	0.030 (0.0032)	0.131 (0.0157)	0.089 (0.0173)	0.096 (0.0133)

[a]Acenaphthene
[b]Acenaphthylene
[c]Anthracene
[d]Benz[a]anthracene
[e]Benzo[a]pyrene
[f]Benzo[b+k]fluoranthene
[g]Benzo[g,h,i]perylene
[h]Chrysene
[i]Fluoranthene
[j]Indeno[1,2,3-cd]pyrene
[k]Naphthalene
[l]Phenanthrene
[m]Pyrene
[n]Not calculated

concentrations below detection limits for acenaphthene, acenaphthylene, anthracene, dibenz[a,h]anthracene, and fluorene. Consequently, 28-day AFs were not calculated for these compounds. In the Hot sediment exposures, AFs were calculated for all PAHs in at least one of the species exposed to both bedded and suspended sediment.

28-day AF comparisons

Twenty-eight-day AF data sets for the bedded sediment exposures were compared statistically with those of the suspended sediment exposures for each organism and sediment combination (TABLE XI). No statistically significant differences were found between 28-day AFs for bedded and suspended sediment exposures for clams in either Reference or Hot sediments, or for mussels in Reference sediments. For mussels exposed to the suspended Hot sediment, 28-day AFs were significantly higher than 28-day AFs for mussels in the bedded Hot sediment exposures ($P \leq 0.05$). These results indicate similar bioavailability of the PAH compounds to clams in suspended as in bedded sediments, and in Hot as in Reference. Similar bioavailability of PAHs to mussels is indicated in Reference bedded as in Reference suspended sediment exposures. However, the 28-day AF means show that the PAHs were twice as available to mussels in the suspended as in the bedded Hot sediment exposures.

TABLE XI

BEDDED VS SUSPENDED SEDIMENT 28-DAY ACCUMULATION FACTOR COMPARISONS

Sediment	Organism	Treatment	n	$\bar{x}$	(SE)	Test Statistic[a]	P
Reference	Mussel (*M. edulis*)	bedded	6	0.162	(0.136)		
		suspended	6	0.171	(0.132)	$X^2 = 0.7811$	0.1884
	Clam (*M. nasuta*)	bedded	10	0.136	(0.103)		
		suspended	10	0.080	(0.139)	$X^2 = 2.2926$	0.0650
Hot	Mussel (*M. edulis*)	bedded	16	0.039	(0.008)		
		suspended	16	0.079	(0.016)	$t = 2.2749$	0.0163[b]
	Clam (*M. nasuta*)	bedded	14	0.042	(0.008)		
		suspended	14	0.043	(0.008)	$X^2 = 0.0085$	0.4634

[a]Test statistic is X^2 when Kruskal-Wallis test is used, t when t-test is used

[b]Significant difference, $\alpha = 0.05$.

COMPARISON OF TBP ESTIMATES WITH MEASURED CONCENTRATIONS

Theoretical Bioaccumulation Potential calculations were made in two ways: (1) using the Green Book recommended AF = 4 (Equation 7), and (2) using the Field-derived AFs for individual PAHs (and for the total of 15 PAHs) shown in TABLE V, in place of the universal AF = 4. The PAH concentration data for the Reference and Hot sediments (TABLE VI), the sediment TOC content, and the lipid content of the bivalves in each treatment (TABLE VIII) were used. Results of the two TBP estimation methods and the concentrations of PAHs measured in organism tissues after 28-day exposures are shown for each treatment in TABLEs XII-XIX and Figures 10-17. A consistent pattern can be seen in which TBP using AF = 4 overestimates the 28-day measured concentrations for both bivalves by one to three orders of magnitude. Conversely, TBP using the field-derived AFs both under- and overestimates the 28-day measured concentrations, but in most cases within one order of magnitude. Pairwise comparisons for the data of TABLEs XII-XIX using LSD, SNK, and Tukey's *W* procedures are shown in TABLE XX for the Reference sediment and TABLE XXI for the Hot sediment. In all cases the TBP (AF = 4) estimations are significantly higher ($P_{\alpha/2} \leq 0.025$) than either the TBP (field AF) estimations or the 28-day tissue concentrations. At the same time, no statistically significant differences are seen

in any treatment between the TBP (field AF) estimations and the 28-day measured tissue concentrations of the PAHs.

The relative similarities and differences between 28-day measured tissue concentrations and the two TBP estimates for the PAHs are summarized in TABLEs XIX (Reference sediment) and XX (Hot sediment). The estimates and the measured concentrations for the four sediment treatments are averaged and ratioed in each TABLE. A ratio of 1.00 would indicate perfect correspondence between estimated and measured concentrations. In the Reference sediment exposures, the TBP estimates using field-AFs range about a factor of 10 above and below the measured concentrations. However, TBP using AF = 4 overestimates in all cases, typically by a few hundred-fold, and in the case of benz[a]anthracene, by nearly three orders of magnitude. In the Hot sediment treatments, TBP estimates using field AFs are highly accurate, ranging about one-half to two-fold the measured concentrations. TBP estimates using AF = 4 are much less accurate, overestimating measured concentrations by factors of 41-386.

TABLE XII

TBP ESTIMATED CONCENTRATIONS AND MEASURED CONCENTRATIONS OF PAH COMPOUNDS IN CLAMS (*M. NASUTA*) EXPOSED FOR 28 DAYS TO BEDDED REFERENCE SEDIMENT

Compound	Concentration, ng g^{-1} wet weight		
	TBP, AF = 4	TBP, Field AF	Measured, 28-day $\bar{x}$ (SE)
Naphthalene	122	3.14	39.3 (10.1)
Acenaphthene	10.3	0.03	1.12 (0.644)
Acenaphthylene	33.6	0.20	NC[a]
Fluorene	10.3	0.01	5.28 (1.39)
Phenanthrene	705	7.67	20.0 (5.16)
Anthracene	173	0.37	0.11 (0.174)
Pyrene	1600	59.1	8.91 (0.916)
Fluoranthene	1537	15.9	8.92 (1.04)
Chrysene	673	10.3	4.25 (0.459)
Benz[a]anthracene	737	7.83	0.73 (0.765)
Benzo[a]pyrene	1226	10.5	4.48 (0.491)
Dibenz[a,h]anthracene	32.3	0.09	0.64 (0.361)
Benzo[b+k]fluoranthene	1413	25.9	9.92 (NC)
Benzo[g,h,i]perylene	813	6.59	4.54 (0.441)
Indeno[1,2,3-cd]pyrene	806	4.53	4.15 (0.305)
Total of 15 PAH	9894	152	112 (NC)

[a]Not calculated.

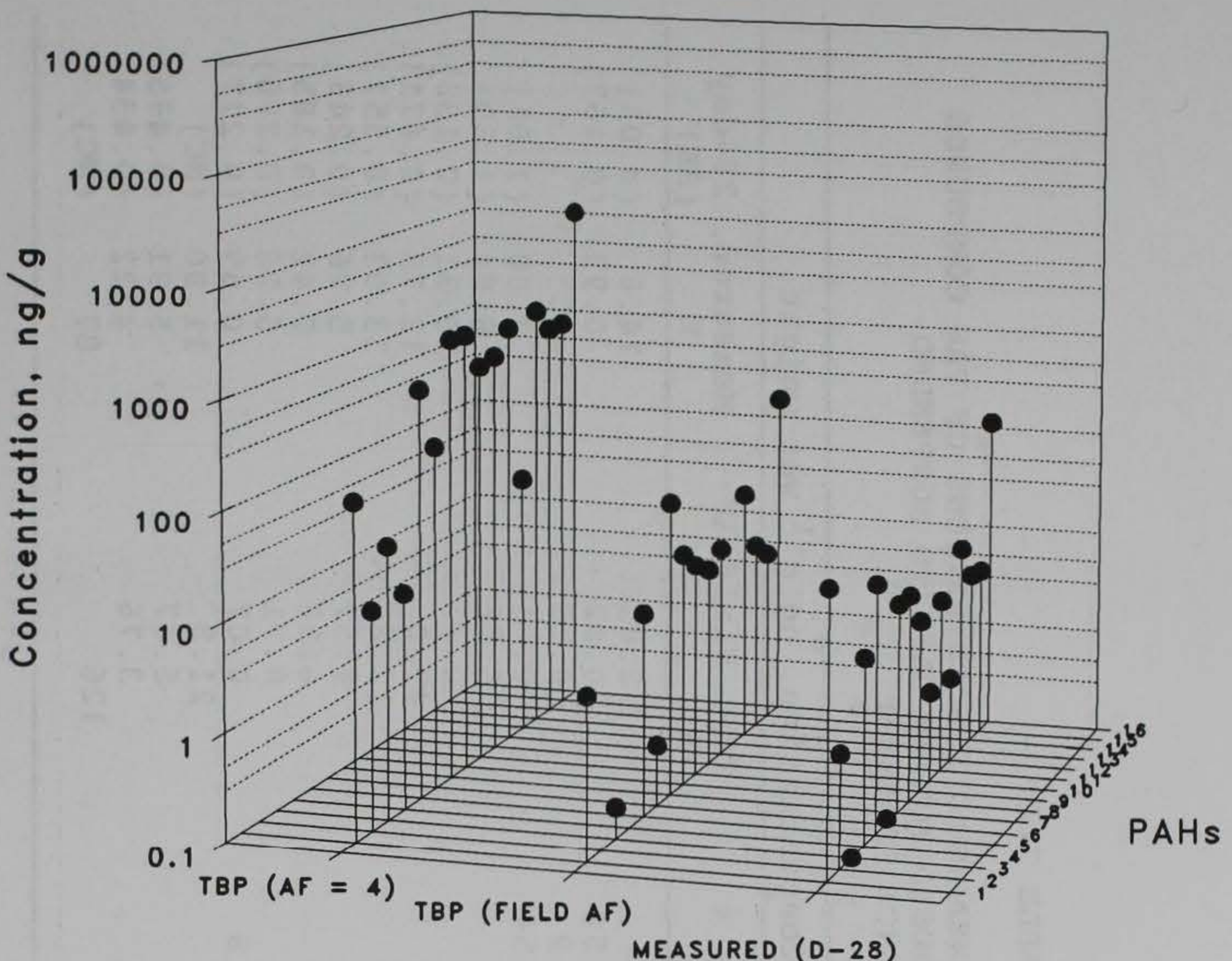


Figure 10. Clams (*M. nasuta*) exposed to bedded Reference sediment. Tissue concentrations of PAH compounds (Measured, D-28), contrasted with TBP estimations made using field-generated AFs (TBP, Field AF) or using Green Book guidance (TBP, AF = 4).

Key to PAHs:

1	naphthalene	9	chrysene
2	acenaphthene	10	benz[a]anthracene
3	acenapthylene	11	benzo[a]pyrene
4	fluorene	12	dibenz[a,h]anthracene
5	phenanthrene	13	benzo[b+k]fluoranthene
6	anthracene	14	benzo[g,h,i,]perylene
7	pyrene	15	indeno[1,2,3-cd]pyrene
8	fluoranthene	16	Total of 15 PAHs

TABLE XIII

TBP ESTIMATED CONCENTRATIONS AND MEASURED CONCENTRATIONS OF PAH COMPOUNDS IN CLAMS (*M. NASUTA*) EXPOSED FOR 28 DAYS TO SUSPENDED REFERENCE SEDIMENT

Compound	Concentration, ng g^{-1} wet weight			
	TBP, AF = 4	TBP, Field AF	Measured, 28-day $\bar{x}$	(SE)
Naphthalene	101	2.61	14.6	(4.03)
Acenaphthene	8.54	0.02	0.93	(0.467)
Acenaphthylene	27.9	0.16	NC[a]	
Fluorene	8.54	0.01	2.00	(1.05)
Phenanthrene	585	6.36	8.84	(1.30)
Anthracene	144	0.31	0.31	(0.133)
Pyrene	1328	49.0	13.75	(0.671)
Fluoranthene	1275	13.2	13.03	(0.551)
Chrysene	559	8.57	5.26	(0.243)
Benz[a]anthracene	611	6.50	0.42	(0.185)
Benzo[a]pyrene	1017	8.71	5.26	(0.296)
Dibenz[a,h]anthracene	26.8	0.07	0.59	(0.217)
Benzo[b+k]fluoranthene	1175	21.5	11.80	(NC)
Benzo[g,h,i]perylene	675	5.47	5.91	(0.475)
Indeno[1,2,3-cd]pyrene	669	3.76	4.22	(0.434)
Total of 15 PAH	8211	126	87	(NC)

[a]Not calculated.

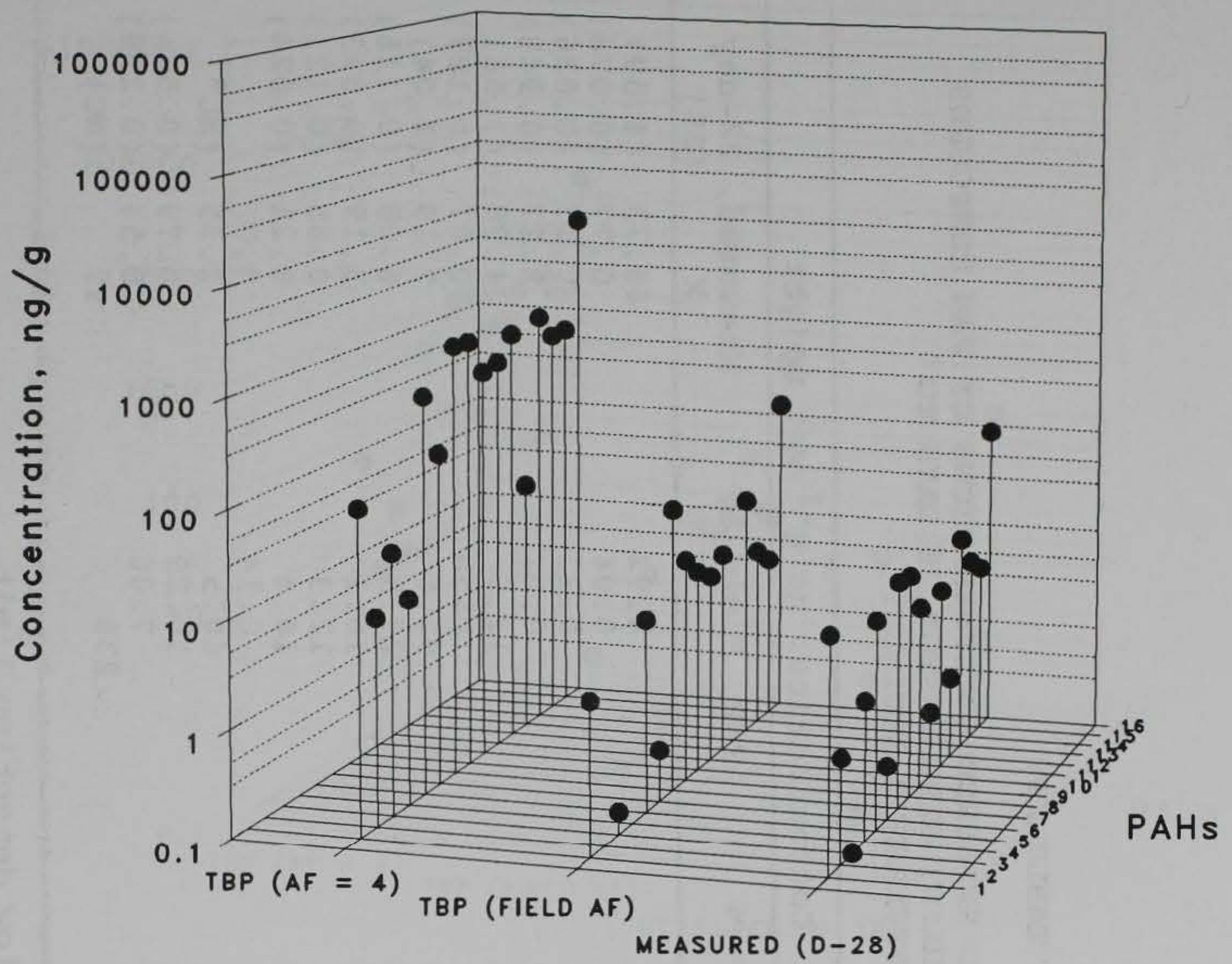


Figure 11. Clams (*M. nasuta*) exposed to continuously suspended Reference sediment. Tissue concentrations of PAH compounds (Measured, D-28), contrasted with TBP estimations made using field-generated AFs (TBP, Field AF) or using Green Book guidance (TBP, AF = 4).

Key to PAHs:

1	naphthalene	9	chrysene
2	acenaphthene	10	benz[a]anthracene
3	acenapthylene	11	benzo[a]pyrene
4	fluorene	12	dibenz[a,h]anthracene
5	phenanthrene	13	benzo[b+k]fluoranthene
6	anthracene	14	benzo[g,h,i,]perylene
7	pyrene	15	indeno[1,2,3-cd]pyrene
8	fluoranthene	16	Total of 15 PAHs

TABLE XIV

TBP-ESTIMATED CONCENTRATIONS AND MEASURED CONCENTRATIONS OF PAH COMPOUNDS IN MUSSELS (*M. EDULIS*) EXPOSED FOR 28 DAYS TO BEDDED REFERENCE SEDIMENT

Compound	Concentration, ng g^{-1} wet weight			
	TBP, AF = 4	TBP, Field AF	Measured, 28-day $\bar{x}$	(SE)
Naphthalene	191	4.92	48.73	(4.59)
Acenaphthene	16.1	0.04	0.77	(0.078)
Acenaphthylene	52.6	0.31	0.17	(0.036)
Fluorene	16.1	0.01	2.27	(0.948)
Phenanthrene	1103	12.0	24.98	(1.56)
Anthracene	271	0.58	0.87	(0.127)
Pyrene	2504	92.5	7.19	(1.04)
Fluoranthene	2404	24.9	6.08	(0.715)
Chrysene	1053	16.2	3.19	(0.373)
Benz[a]anthracene	1153	12.3	0.65	(0.121)
Benzo[a]pyrene	1918	16.4	0.21	(0.024)
Dibenz[a,h]anthracene	50.6	0.14	0.0	
Benzo[b+k]fluoranthene	2216	40.5	2.35	(NC[a])
Benzo[g,h,i]perylene	1272	10.29	0.79	(0.685)
Indeno[1,2,3-cd]pyrene	1262	7.08	0.54	(0.228)
Total of 15 PAH	15480	238	99	(NC)

[a]Not available. All samples below detection limit.

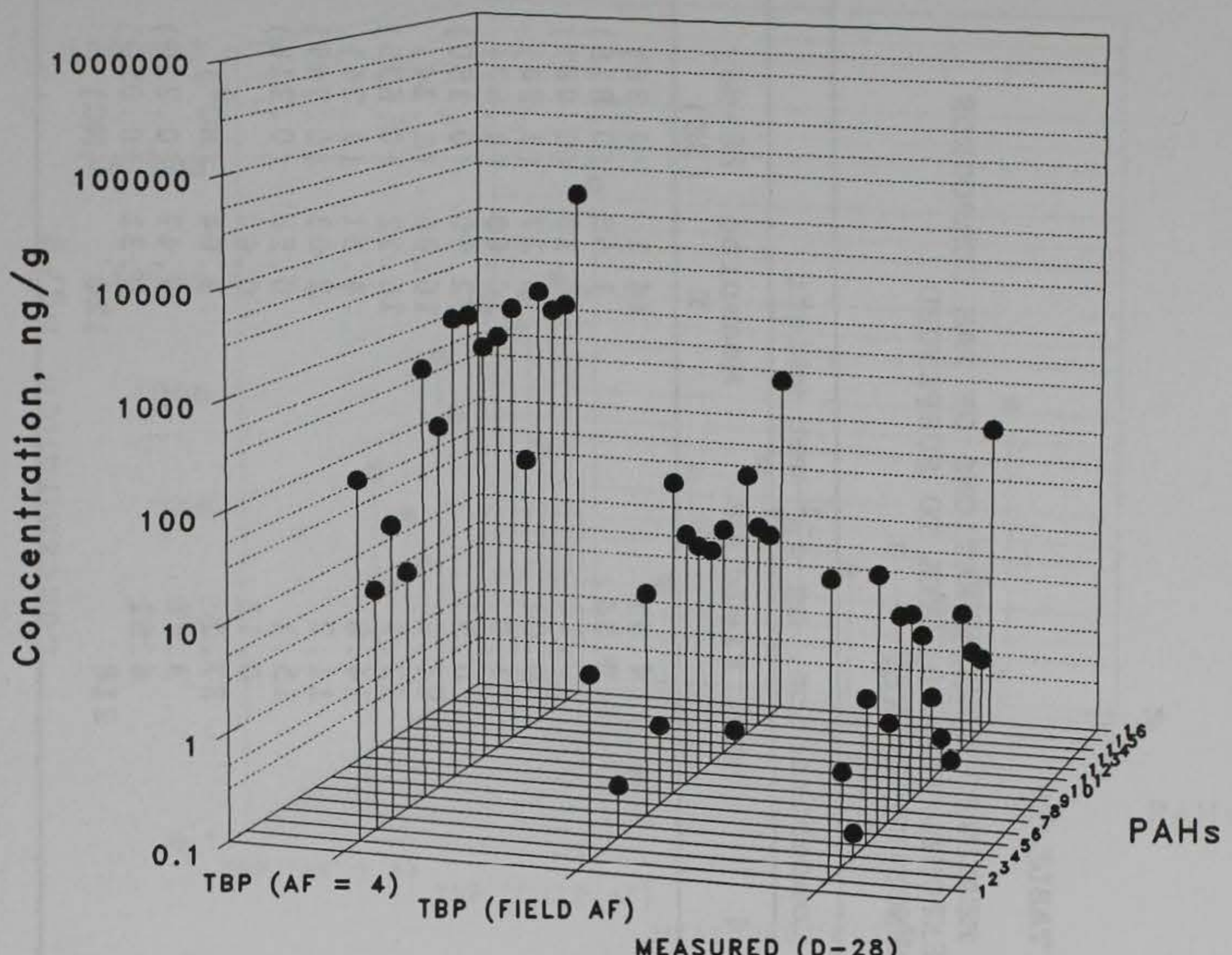


Figure 12. Mussels (*M. edulis*) exposed to bedded Reference sediment. Tissue concentrations of PAH compounds (Measured, D-28), contrasted with TBP estimations made using field-generated AFs (TBP, Field AF) or using Green Book guidance (TBP, AF = 4).

Key to PAHs:

1	naphthalene	9	chrysene
2	acenaphthene	10	benz[a]anthracene
3	acenapthylene	11	benzo[a]pyrene
4	fluorene	12	dibenz[a,h]anthracene
5	phenanthrene	13	benzo[b+k]fluoranthene
6	anthracene	14	benzo[g,h,i,]perylene
7	pyrene	15	indeno[1,2,3-cd]pyrene
8	fluoranthene	16	Total of 15 PAHs

TABLE XV

TBP-ESTIMATED CONCENTRATIONS AND MEASURED CONCENTRATIONS OF PAH COMPOUNDS IN MUSSELS (*M. EDULIS*) EXPOSED FOR 28 DAYS TO SUSPENDED REFERENCE SEDIMENT

Compound	Concentration, ng g^{-1} wet weight			
	TBP, AF = 4	TBP, Field AF	Measured, 28-day $\bar{x}$	(SE)
Naphthalene	175	4.51	44.1	(4.36)
Acenaphthene	14.8	0.04	1.65	(0.816)
Acenaphthylene	48.2	0.28	0.13	(0.021)
Fluorene	14.8	0.01	3.27	(1.24)
Phenanthrene	1012	11.0	30.60	(1.87)
Anthracene	249	0.53	2.50	(0.150)
Pyrene	2297	84.8	16.00	(1.34)
Fluoranthene	2206	22.9	10.32	(0.635)
Chrysene	966	14.8	4.51	(1.26)
Benz[a]anthracene	1057	11.2	2.03	(0.749)
Benzo[a]pyrene	1759	15.1	0.58	(0.218)
Dibenz[a,h]anthracene	46.4	0.13	0.0	
Benzo[b+k]fluoranthene	2033	37.1	3.64	(NC[a])
Benzo[g,h,i]perylene	1167	9.46	0.47	(0.225)
Indeno[1,2,3-cd]pyrene	1158	6.51	0.31	(0.050)
Total of 15 PAH	14201	218	120	(NC)

[a]Not calculated.

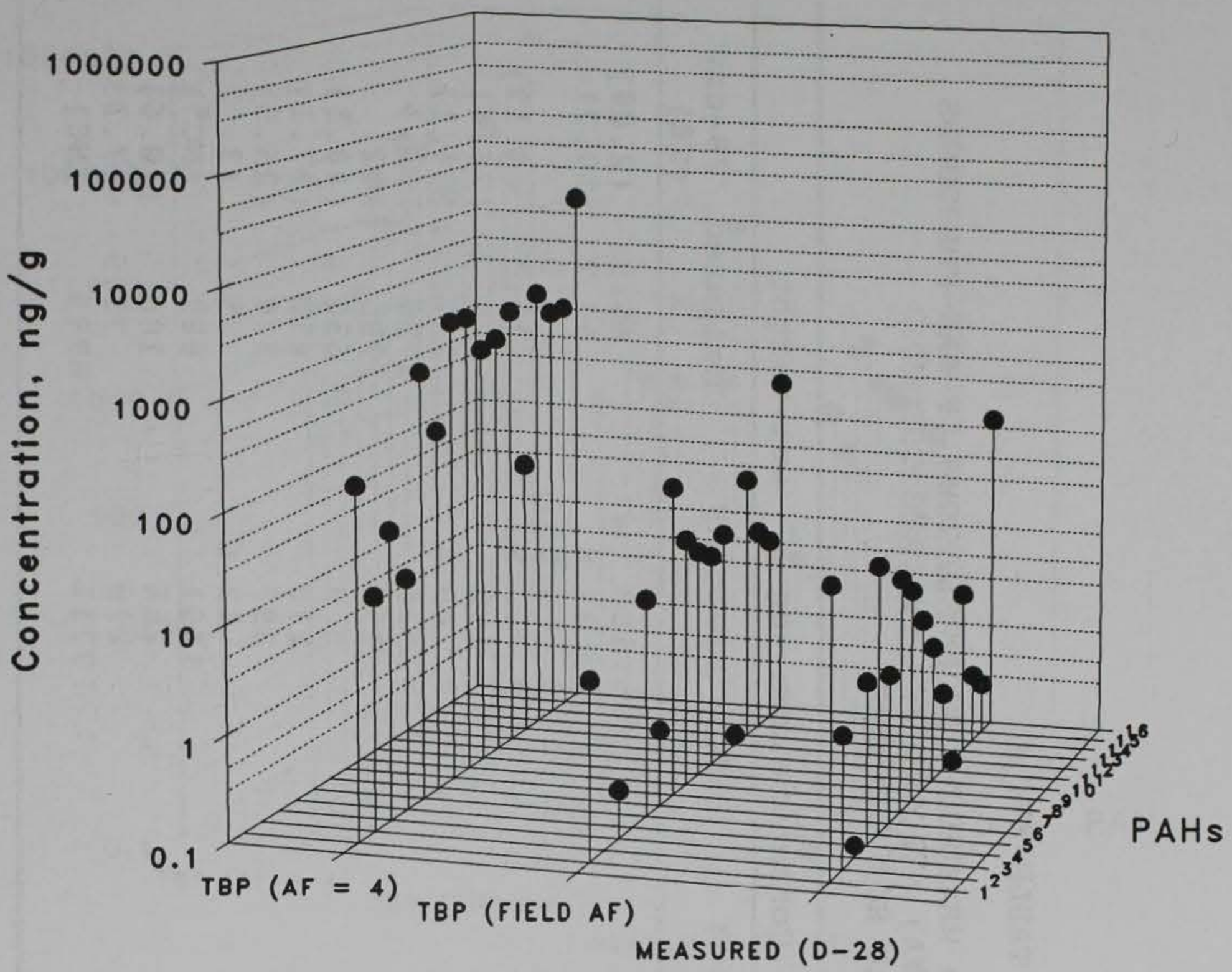


Figure 13. Mussels (*M. edulis*) exposed to continuously suspended Reference sediment. Tissue concentrations of PAH compounds (Measured, D-28), contrasted with TBP estimations made using field-generated AFs (TBP, Field AF) or using Green Book guidance (TBP, AF = 4).

Key to PAHs:

1	naphthalene	9	chrysene
2	acenaphthene	10	benz[a]anthracene
3	acenapthylene	11	benzo[a]pyrene
4	fluorene	12	dibenz[a,h]anthracene
5	phenanthrene	13	benzo[b+k]fluoranthene
6	anthracene	14	benzo[g,h,i,]perylene
7	pyrene	15	indeno[1,2,3-cd]pyrene
8	fluoranthene	16	Total of 15 PAHs

TABLE XVI

TBP-ESTIMATED CONCENTRATIONS AND MEASURED CONCENTRATIONS OF PAH COMPOUNDS IN CLAMS (*M. NASUTA*) EXPOSED FOR 28 DAYS TO BEDDED HOT SEDIMENT

Compound	Concentration, ng g^{-1} wet weight			
	TBP, AF = 4	TBP, Field AF	Measured, 28-day $\bar{x}$	(SE)
Naphthalene	5926	153	65.9	(2.08)
Acenaphthene	13350	34.8	53.2	(10.7)
Acenaphthylene	747	4.35	0.0	
Fluorene	5541	3.93	32.3	(4.13)
Phenanthrene	54445	592	634	(74.4)
Anthracene	19028	40.4	237	(24.1)
Pyrene	78979	2916	1750	(102)
Fluoranthene	76738	796	1785	(112)
Chrysene	34522	530	630	(40.5)
Benz[a]anthracene	25956	276	452	(27.1)
Benzo[a]pyrene	46396	397	320	(25.5)
Dibenz[a,h]anthracene	4676	12.6	25	(3.12)
Benzo[b+k]fluoranthene	79389	1451	629	(NC[a])
Benzo[g,h,i]perylene	35212	286	100	(10.6)
Indeno[1,2,3-cd]pyrene	38789	218	71.9	(7.33)
Total of 15 PAH	520603	7711	6785	(NC)

[a]Not calculated.

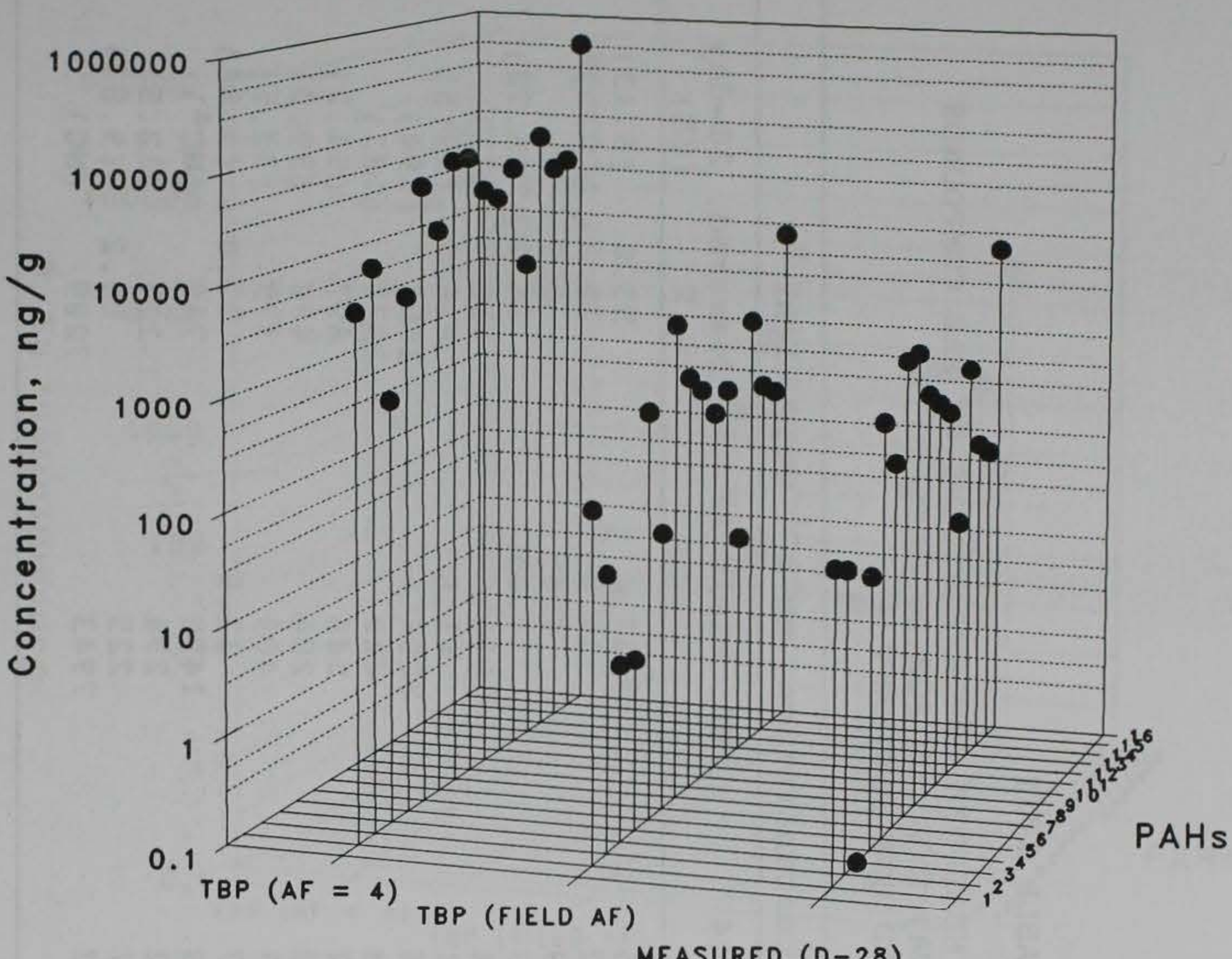


Figure 14. Clams (*M. nasuta*) exposed to bedded Hot sediment. Tissue concentrations of PAH compounds (Measured, D-28), contrasted with TBP estimations made using field-generated AFs (TBP, Field AF) or using Green Book guidance (TBP, AF = 4).

Key to PAHs:

1	naphthalene	9	chrysene
2	acenaphthene	10	benz[a]anthracene
3	acenapthylene	11	benzo[a]pyrene
4	fluorene	12	dibenz[a,h]anthracene
5	phenanthrene	13	benzo[b+k]fluoranthene
6	anthracene	14	benzo[g,h,i,]perylene
7	pyrene	15	indeno[1,2,3-cd]pyrene
8	fluoranthene	16	Total of 15 PAHs

TABLE XVII

TBP-ESTIMATED CONCENTRATIONS AND MEASURED CONCENTRATIONS OF PAH COMPOUNDS IN CLAMS (*M. NASUTA*) EXPOSED FOR 28 DAYS TO SUSPENDED HOT SEDIMENT

Compound	Concentration, ng g^{-1} wet weight			
	TBP, AF = 4	TBP, Field AF	Measured, 28-day $\bar{x}$	(SE)
Naphthalene	6105	157	53.6	(6.13)
Acenaphthene	13752	35.8	39.6	(10.5)
Acenaphthylene	769	4.48	0.0	
Fluorene	5927	4.05	24.9	(5.72)
Phenanthrene	56084	610	610	(111)
Anthracene	19601	41.6	216	(39.5)
Pyrene	81356	3004	1958	(194)
Fluoranthene	79048	820	1871	(181)
Chrysene	35562	546	697	(54.2)
Benz[a]anthracene	26738	284	492	(39.8)
Benzo[a]pyrene	47793	409	379	(33.5)
Dibenz[a,h]anthracene	4817	13.0	27.0	(1.84)
Benzo[b+k]fluoranthene	81778	1495	719	(NC[a])
Benzo[g,h,i]perylene	36272	294	117	(12.5)
Indeno[1,2,3-cd]pyrene	39957	225	85.2	(9.83)
Total of 15 PAH	535557	7943	7289	(NC)

[a]Not calculated.

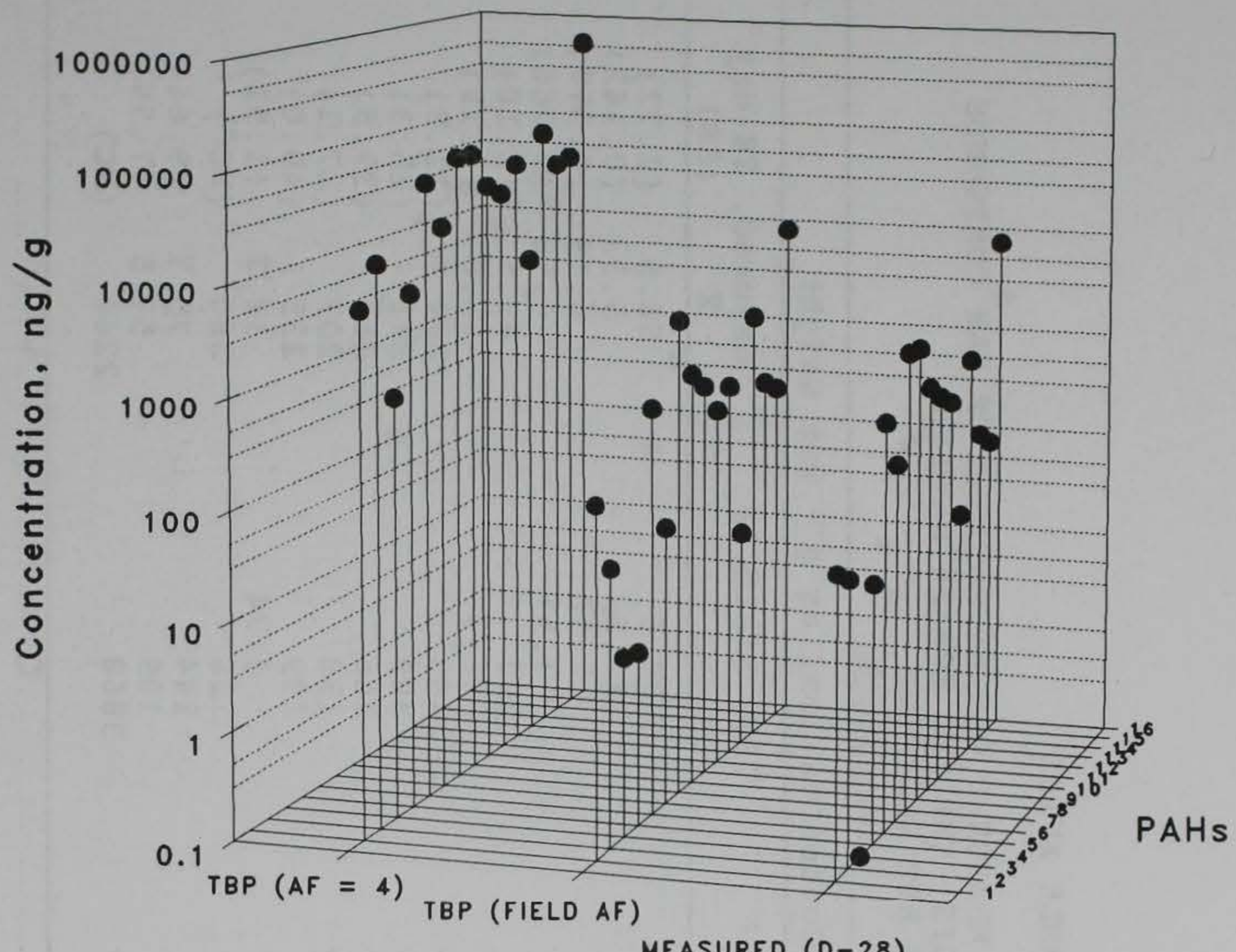


Figure 15. Clams (*M. nasuta*) exposed to continuously suspended Hot sediment. Tissue concentrations of PAH compounds (Measured, D-28), contrasted with TBP estimations made using field-generated AFs (TBP, Field AF) or using Green Book guidance (TBP, AF = 4).

Key to PAHs:

1	naphthalene	9	chrysene
2	acenaphthene	10	benz[a]anthracene
3	acenapthylene	11	benzo[a]pyrene
4	fluorene	12	dibenz[a,h]anthracene
5	phenanthrene	13	benzo[b+k]fluoranthene
6	anthracene	14	benzo[g,h,i,]perylene
7	pyrene	15	indeno[1,2,3-cd]pyrene
8	fluoranthene	16	Total of 15 PAHs

TABLE XVIII

TBP-ESTIMATED CONCENTRATIONS AND MEASURED CONCENTRATIONS OF PAH COMPOUNDS IN MUSSELS (*M. EDULIS*) EXPOSED FOR 28 DAYS TO BEDDED HOT SEDIMENT

Compound	Concentration, ng g^{-1} wet weight			
	TBP, AF = 4	TBP, Field AF	Measured, 28-day $\bar{x}$	(SE)
Naphthalene	2973	76.6	58.5	(1.72)
Acenaphthene	6697	17.5	5.33	(0.161)
Acenaphthylene	375	2.18	6.19	(0.577)
Fluorene	2887	1.97	4.58	(0.066)
Phenanthrene	27314	297	49.5	(2.10)
Anthracene	9546	20.3	13.8	(1.19)
Pyrene	39622	1463	294	(21.3)
Fluoranthene	38497	399	311	(25.3)
Chrysene	17319	266	361	(39.5)
Benz[a]anthracene	13022	138	207	(27.1)
Benzo[a]pyrene	23276	199	235	(40.0)
Dibenz[a,h]anthracene	2346	6.34	12.3	(1.91)
Benzo[b+k]fluoranthene	39827	728	560	(NC[a])
Benzo[g,h,i]perylene	17665	144	75.3	(9.64)
Indeno[1,2,3-cd]pyrene	19460	109	51.5	(7.56)
Total of 15 PAH	260823	3868	2244	(NC)

[a]Not calculated.

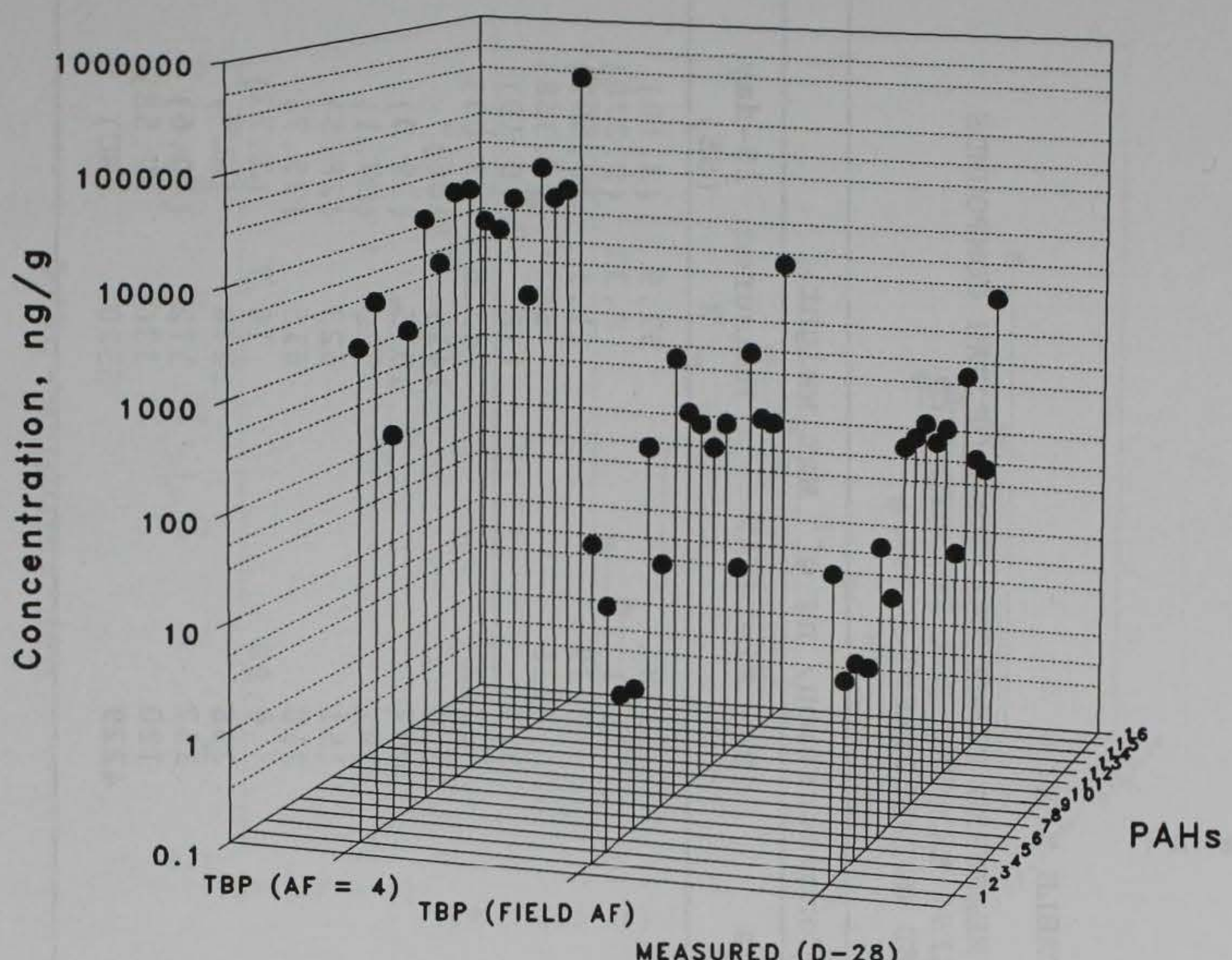


Figure 16. Mussels (*M. edulis*) exposed to bedded Hot sediment. Tissue concentrations of PAH compounds (Measured, D-28), contrasted with TBP estimations made using field-generated AFs (TBP, Field AF) or using Green Book guidance (TBP, AF = 4).

Key to PAHs:

1	naphthalene	9	chrysene
2	acenaphthene	10	benz[a]anthracene
3	acenapthylene	11	benzo[a]pyrene
4	fluorene	12	dibenz[a,h]anthracene
5	phenanthrene	13	benzo[b+k]fluoranthene
6	anthracene	14	benzo[g,h,i,]perylene
7	pyrene	15	indeno[1,2,3-cd]pyrene
8	fluoranthene	16	Total of 15 PAHs

TABLE XIX

TBP-ESTIMATED CONCENTRATIONS AND MEASURED CONCENTRATIONS OF PAH COMPOUNDS IN MUSSELS (*M. EDULIS*) EXPOSED FOR 28 DAYS TO SUSPENDED HOT SEDIMENT

Compound	Concentration, ng g^{-1} wet weight			
	TBP, AF = 4	TBP, Field AF	Measured, 28-day $\bar{x}$	(SE)
Naphthalene	3250	83.8	57.9	(3.10)
Acenaphthene	7322	19.1	8.33	(0.378)
Acenaphthylene	410	2.39	11.1	(0.597)
Fluorene	3156	2.16	6.77	(0.368)
Phenanthrene	29863	325	138	(8.19)
Anthracene	10437	22.2	42.5	(2.03)
Pyrene	43320	1600	1415	(106)
Fluoranthene	42090	437	1075	(39.0)
Chrysene	18935	291	925	(37.1)
Benz[a]anthracene	14237	151	623	(28.5)
Benzo[a]pyrene	25448	218	615	(35.7)
Dibenz[a,h]anthracene	2565	6.93	16.7	(3.77)
Benzo[b+k]fluoranthene	43544	796	1278	(NC[a])
Benzo[g,h,i]perylene	19314	157	172	(10.6)
Indeno[1,2,3-cd]pyrene	21276	120	136	(9.58)
Total of 15 PAH	285167	4229	6520	(NC)

[a]Not calculated.

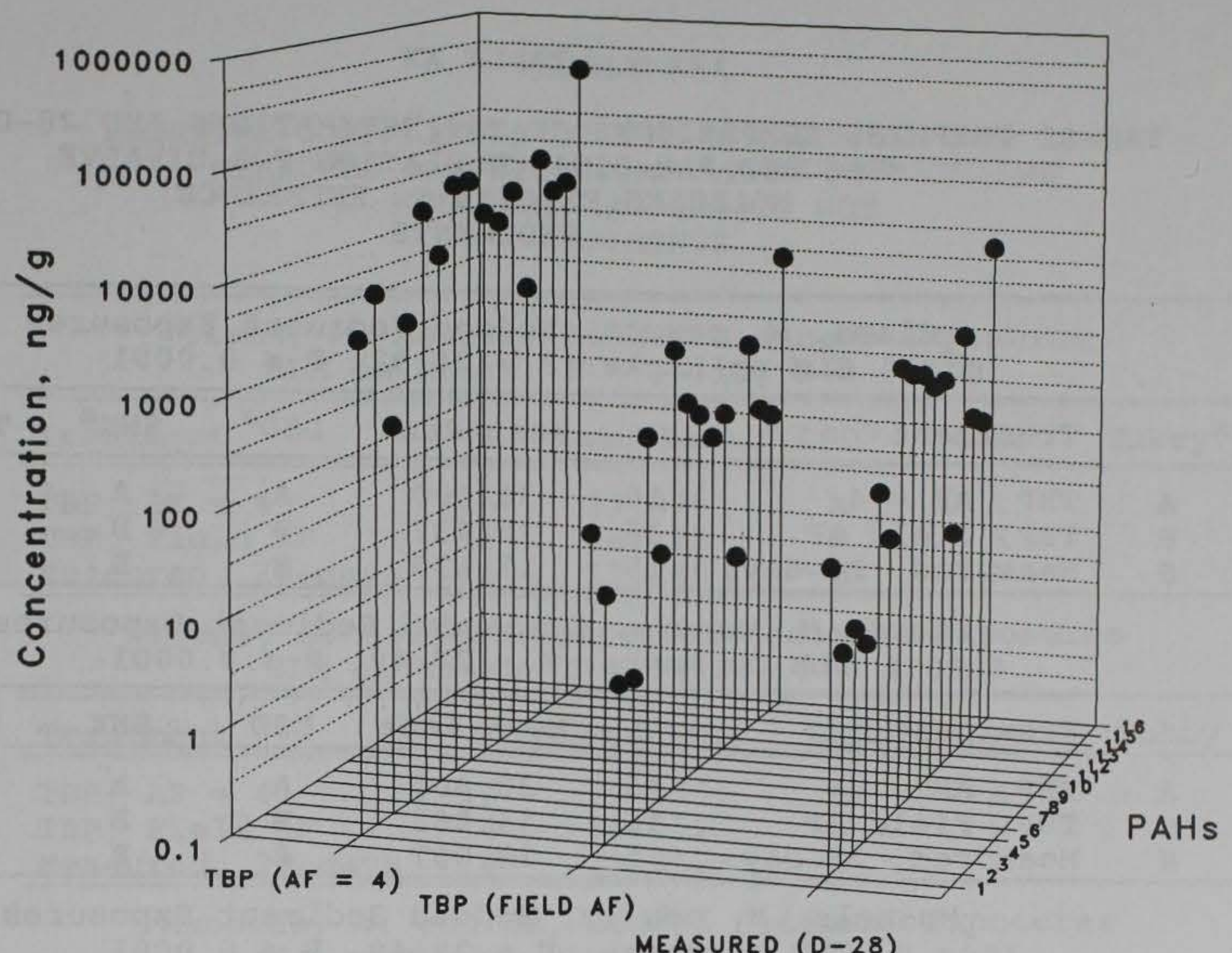


Figure 17. Mussels (*M. edulis*) exposed to continuously suspended Hot sediment. Tissue concentrations of PAH compounds (Measured, D-28), contrasted with TBP estimations made using field-generated AFs (TBP, Field AF) or using Green Book guidance (TBP, AF = 4).

Key to PAHs:

1	naphthalene	9	chrysene
2	acenaphthene	10	benz[a]anthracene
3	acenapthylene	11	benzo[a]pyrene
4	fluorene	12	dibenz[a,h]anthracene
5	phenanthrene	13	benzo[b+k]fluoranthene
6	anthracene	14	benzo[g,h,i,]perylene
7	pyrene	15	indeno[1,2,3-cd]pyrene
8	fluoranthene	16	Total of 15 PAHs

TABLE XX

PAIRWISE COMPARISONS OF TBP ESTIMATIONS AND 28-DAY MEASURED PAH BIOACCUMULATION FOR BIVALVE MOLLUSKS EXPOSED TO REFERENCE SEDIMENTS

Clams, *M. nasuta*, Bedded Sediment Exposures
GLM on Ranks: F = 26.92, P ≤ 0.0001

Treatment	*n*	Mean Rank	LSD[a]	SNK[a]	Tukey[a]
TBP, AF = 4	16	38.000	A	A	A
TBP, Field AF	16	16.437	B	B	B
Measured, 28-days	15	17.133	B	B	B

Clams, *M. nasuta*, Suspended Sediment Exposures
GLM on Ranks: F = 27.49, P ≤ 0.0001

Treatment	*n*	Mean Rank	LSD	SNK	Tukey
TBP, AF = 4	16	38.000	A	A	A
TBP, Field AF	16	15.563	B	B	B
Measured, 28-days	15	18.067	B	B	B

Mussels, *M. edulis*, Bedded Sediment Exposures
GLM on Ranks: F = 29.40, P ≤ 0.0001

Treatment	*n*	Mean Rank	LSD	SNK	Tukey
TBP, AF = 4	16	39.125	A	A	A
TBP, Field AF	16	15.563	B	B	B
Measured, 28-days	16	18.812	B	B	B

Mussels, *M. edulis*, Suspended Sediment Exposures
GLM on Ranks: F = 27.56, P ≤ 0.0001

Treatment	*n*	Mean Rank	LSD	SNK	Tukey
TBP, AF = 4	16	39.000	A	A	A
TBP, Field AF	16	16.375	B	B	B
Measured, 28-days	16	18.125	B	B	B

[a]Treatments with same letter designation do not differ significantly, $P_{\alpha/2} \leq 0.025$.

TABLE XXI

PAIRWISE COMPARISONS OF TBP ESTIMATIONS AND 28-DAY MEASURED PAH BIOACCUMULATION FOR BIVALVE MOLLUSKS EXPOSED TO HOT SEDIMENTS

Clams, *M. nasuta*, Bedded Sediment Exposures
GLM on Ranks: F = 36.18, P ≤ 0.0001

Treatment	*n*	Mean Rank	LSD[a]	SNK[a]	Tukey[a]
TBP, AF = 4	16	39.875	A	A	A
TBP, Field AF	16	16.375	B	B	B
Measured, 28-days	16	17.250	B	B	B

Clams, *M. nasuta*, Suspended Sediment Exposures
GLM on Ranks: F = 36.21, P ≤ 0.0001

Treatment	*n*	Mean Rank	LSD	SNK	Tukey
TBP, AF = 4	16	39.875	A	A	A
TBP, Field AF	16	16.312	B	B	B
Measured, 28-days	16	17.312	B	B	B

Mussels, *M. edulis*, Bedded Sediment Exposures
GLM on Ranks: F = 40.188, P ≤ 0.0001

Treatment	*n*	Mean Rank	LSD	SNK	Tukey
TBP, AF = 4	16	40.188	A	A	A
TBP, Field AF	16	16.062	B	B	B
Measured, 28-days	16	17.250	B	B	B

Mussels, *M. edulis*, Suspended Sediment Exposures
GLM on Ranks: F = 34.69, P ≤ 0.0001

Treatment	*n*	Mean Rank	LSD	SNK	Tukey
TBP, AF = 4	16	39.750	A	A	A
TBP, Field AF	16	16.469	B	B	B
Measured, 28-days	16	17.281	B	B	B

[a]Treatments with same letter designation do not differ significantly, $P_{\alpha/2} \leq 0.025$.

TABLE XXII

RATIOS OF TBP ESTIMATE MEANS[a] TO 28-DAY MEASURED TISSUE CONCENTRATION MEANS FOR THE REFERENCE SEDIMENT

Compound	TBP-Field/Day-28[b]	TBP-AF4/Day-28[c]
Naphthalene	0.10	4.0
Acenaphthene	0.03	11
Acenaphthylene	1.60	274
Fluorene	0.00	3.9
Phenanthrene	0.44	40
Anthracene	0.47	220
Pyrene	6.23	169
Fluoranthene	2.01	194
Chrysene	2.90	189
Benz[a]anthracene	9.88	930
Benzo[a]pyrene	4.82	562
Dibenz[a,h]anthracene	0.34	126
Benzo[b+k]fluoranthene	4.51	247
Benzo[g,h,i]perylene	2.72	335
Indeno[1,2,3-cd]pyrene	2.37	422
Total of 15 PAH	1.76	115

[a]Average of the four sediment treatments.

[b]Ratio of average estimate using field-derived AFs to average measured concentration.

[c]Ratio of average estimate using AF = 4 to average measured concentration.

TABLE XXIII

RATIOS OF TBP ESTIMATE MEANS[a] TO 28-DAY MEASURED TISSUE CONCENTRATION MEANS FOR THE HOT SEDIMENT

Compound	TBP-Field/Day-28[b]	TBP-AF4/Day-28[c]
Naphthalene	1.99	77
Acenaphthene	1.01	386
Acenaphthylene	0.78	133
Fluorene	0.18	259
Phenanthrene	1.27	117
Anthracene	0.24	115
Pyrene	1.66	45
Fluoranthene	0.49	47
Chrysene	0.62	41
Benz[a]anthracene	0.48	45
Benzo[a]pyrene	0.79	92
Dibenz[a,h]anthracene	0.48	179
Benzo[b+k]fluoranthene	1.40	77
Benzo[g,h,i]perylene	1.89	234
Indeno[1,2,3-cd]pyrene	1.95	347
Total of 15 PAH	1.04	70

[a]Average of the four sediment treatments.

[b]Ratio of average estimate using field-derived AFs to average measured concentration.

[c]Ratio of average estimate using AF = 4 to average measured concentration.

DISCUSSION

TRACE CONTAMINANT DATA

The location chosen for sampling benthic organisms and sediments in the New York Bight Apex is used as a source of reference sediments for the conduct of Green Book bioassays and bioaccumulation tests for dredged material disposal evaluations in New York and Newark, NJ, Harbors. The sampling area is considered to be relatively free of contamination, and the results of this investigation showed that concentrations of PAH compounds in sediments and benthic organisms at the site are generally low.

Concentrations of individual PAH compounds in invertebrate organisms collected from recognized contaminated sites have been measured at tens of parts-per-million. For example, benzo[a]pyrene and benzo[b]-fluoranthene were reported as 21 and 56 ppm, respectively, in mussels from a coastal area of Southern California (Dunn and Young 1976). The organisms collected at the New York Bight Apex field site range three orders of magnitude lower and are clearly not contaminated by comparison. Some of the analytes (e.g., acenaphthene, acenaphthylene, fluorene, and anthracene) were frequently reported below DL, and a large measure of uncertainty is associated with the concentrations calculated for these compounds. Data near DL can be greatly influenced by random variability or instrument "noise."

These data are inherently less reliable than values quantitated well above the DL.

Data of trace chemical analyses reported below DL are frequently encountered in environmental assessments and present difficulties for interpretation. If such data are simply discarded, they can represent a substantial loss in information and resources. Additionally, poor estimates of statistical parameters result, and the probability of an erroneous outcome in statistical comparisons is increased by the elimination of nondetects (Clarke 1992, Clarke and Brandon 1994). The conventions described above for handling data reported as less than DL were selected, based on past experience with similar data sets, as the best interim practice pending development of statistical guidance. Guidance based on Monte Carlo simulations involving several hundred sediment and biota trace chemical analysis data sets is currently under development at the USAE WES (Clarke and Brandon 1994).

BIOACCUMULATION

<u>PAHs in fish</u>

Because fish generally have a well developed metabolic capability for detoxication of foreign compounds, and because unsubstituted PAH compounds are particularly labile, the failure to detect tissue residues of these compounds in the flatfish exposed with the two bivalves to the Reference and Hot sediments does not mean they were not bioavailable.

Numerous studies have reported high incidences of tumors and lesions in fish from areas of high PAH contamination (Malins et al. 1984, 1987, 1988; Roubal and Malins 1985, Mix et al. 1986). Malins et al. (1984) found consistent positive correlations between the prevalence of hepatic neoplasms and lesions in English sole and sculpin, and high concentrations of PAH compounds in sediments of Puget sound. Similar positive correlations were not found for chlorinated organic compounds detected in sediments and as residues in fish.

In addition, the cancerous effects were found to be consistent with metabolic studies in organisms exposed to benzo[a]pyrene. More of the ultimate carcinogenic metabolites of benzo[a]pyrene (B[a]P-7,8-diol-9,10-epoxides) were produced and greater covalent binding of the metabolites occurred in sole liver than in rat liver in laboratory studies, indicating a greater potential of the fish for development of cancer from PAH exposure than of the rat (Stein et al. 1984).

The bioavailability of these compounds to flatfish from contaminated sediments and the metabolic disposition that follows has been amply demonstrated in the laboratory using radiotracer techniques (Stein et al.1984, 1987). Exposure of various species of fish in the laboratory to PAH compounds that are known human or other mammalian carcinogens, e.g., benzo[a]pyrene and 7,12-dimethylbenz[a]anthracene, have been demonstrated to cause

the development of cancers similar to those observed in fish collected from contaminated areas (Black et al. 1988; Metcalfe et al. 1988; Hawkins et al. 1989).

PAHs in Bivalves

Although bivalve mollusks are not devoid of the ability to metabolize PAH compounds, their ability to do so is much less than that of fish and other vertebrate organisms. Consequently, most bivalves will bioaccumulate PAHs as the parent compound, and do not as readily show the same toxic effects (tumors, lesions, tissue death and necrosis, etc.) caused by reactive metabolites of PAH compounds and typically seen in fish exposed to these compounds.

For example, *Mytilus galloprovincialis* and *M. edulis* exposed in the laboratory to PAHs showed increases in cytochrome P-450 content and NADPH cytochrome c reductase activity but did not show a concomitant increase in benzo[a]pyrene hydroxylase activity (BPH) (Gilewicz et al. 1984, Livingstone and Farrar 1985, Livingstone et al. 1986). Stegeman (1985) reported BPH activity in *M. edulis*, but the results were inconsistent, appeared to be seasonal, and were thought possibly to involve other catalytic processes. Evidently, although the bivalves possess a mixed function oxidase (MFO), oxidative metabolic biotransformation system, the system does not include the enzymes required for metabolism of PAH compounds to an appreciable extent.

Effect of feeding type

Several laboratory studies have been reported in which both suspension-feeding and deposit-feeding bivalve mollusks have been exposed to radiolabeled PAH compounds dosed in sediments, water, and food (Roesijadi et al. 1978; Augenfeld and Anderson 1982, Fortner and Sick 1985; Foster et al. 1987). PAH compounds were readily bioaccumulated by all routes of administration and rates and levels achieved generally related to relative hydrophobicity similarly to the chlorinated hydrocarbons. Comparisons of feeding types showed deposit feeders bioaccumulating to higher levels than suspension feeders (Roesijadi et al. 1978). These authors did not find naphthalene bioaccumulating in *Protothaca staminea*, whereas more hydrophobic alkylated naphthalenes showed detectable amounts in this filter-feeding clam after 60 days. The deposit-feeding clam, *Macoma inquinata*, readily bioaccumulated all PAH compounds. Similar results were obtained by Foster et al. (1987) with another filter-feeding clam, *Mya arenaria*, and a deposit-feeder, *Macoma balthica*.

The 28-day measured tissue concentrations (TABLEs XII-XIX) and the 28-day AF comparisons in TABLE XI show that the suspension-feeding mussel frequently bioaccumulated individual PAH compounds to nearly the same level, and in some cases to higher levels, than the deposit feeding clam. These results indicate clearly that, given similar

lipoidicities, far more than feeding-type is involved as a determinant of potential for bioaccumulation. Rather, the influence of feeding-type is relevant only with reference to the contaminant fugacity in the contaminant source.

Implications for PAH bioavailability

In the experiments described here, high and constant levels of contaminated sediments suspended in the water column provided *M. nasuta* with virtually the same exposure as did the same material when it was bedded. This is evidenced by the similarity of the mean 28-day AFs for the clam in the Hot bedded and suspended sediment exposures (0.042 and 0.043, respectively) (TABLE XI). The mean 28-day AFs for the clams in the Reference exposures are not as nearly identical, but nevertheless, are not statistically different. Clearly, the ability of *M. nasuta* to both filter- and deposit-feed provides it with equivalent exposures. By contrast, the mean 28-day AFs of the mussel are statistically different in the Hot bedded and suspended sediment exposures. The mussels bioaccumulated twice as much from the suspended Hot material, which they actively filter from the water, than from the bedded, which must desorb PAHs to the overlying water in order for these compounds to be available for uptake. The same trend is not seen in the Reference sediments, which are far less contaminated.

The intensity of exposures to suspended sediment-associated contaminants under these conditions is not intended to represent exposures in full-scale aquatic systems. However, the results of the study indicate a potential for bioavailability of PAHs from suspended sediments that is comparable to that from bedded sediment, and in the case of filter-feeding organisms not normally in contact with bedded sediment, the potential appears to be greater when contaminant loading is high.

The interpretation that can be given to the similarities in bioaccumulation shown by the mean 28-day accumulation factors for mussels and clams in suspended and bedded exposures is that bioavailability of PAH compounds was largely the same throughout an exposure aquarium. The significantly greater bioavailability of PAHs to the mussels in the suspended Hot exposure as compared with the bedded Hot exposure can be explained by the feeding habit of the mussels, which involves a very high filtration rate and the processing and concentration of suspended particulate matter. The implication is that fish exposed with the bivalves received comparable exposures. The presence of the higher molecular weight PAHs as parent compounds in the bivalve mollusks and not in the flatfish is consistent with these observations. The inference is that the fish metabolically degraded the PAH compounds. This conclusion is also consistent with the fact that among the PAHs only

naphthalene and phenanthrene were present in fish tissues at appreciable levels. It was previously reported that naphthalene and phenanthrene can both bioaccumulate as the parent compound in fish, whereas the other PAH compounds do not (Gerhart *et al*. 1981, McCarthy and Jimenez 1985). In practical terms, these observations support the utility of benthic clams, *M. nasuta*, and fouling community mussels, *M. edulis*, as sentinel organisms for PAH pollutant monitoring in estuaries.

PHYSICAL PROPERTIES, TISSUE RESIDUES, AND TOXICITY

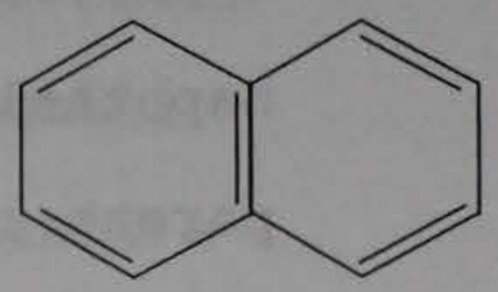
Naphthalene, acenaphthene, and acenaphthylene

Structures of the three compounds are shown in Figure 18. The three share the same fused two-phenyl ring nucleus (i.e., naphthalene) with acenaphthene and acenaphthylene having a two-carbon bridge across the phenyls forming a third ring. The more saturated compound, acenaphthene, has the lowest water solubility of the three, and although slightly less planar, would be expected to bioaccumulate to higher levels in the absence of metabolism. Of the three, only acenaphthene has shown mutagenicity in the Ames Test (cited in Verschueren 1983). Naphthalene did not bioaccumulate to significant levels in either bivalve under any conditions of exposure to Reference (Figures 2-5) or Hot (Figures 6-9) sediments. *M. edulis* bioaccumulated 5.33-11.1 ng g^{-1} acenaphthene and acenaphthylene from bedded and suspended Hot sediments, and *M. nasuta* bioaccumulated

acenaphthene in bedded Hot sediment exposures to a high of 53.2 ng g^{-1}, but did not bioaccumulate acenaphthylene.

In benthic mollusks and polychaetes collected in sediment grab samples in the New York Bight Apex field study, naphthalene was measured at similar concentrations (20.6-34.6 ng g^{-1}) in all species, regardless of taxa or lipid content. Acenaphthylene was found in the mollusks, *Nucula sp.*, 7.06 ng g^{-1}, and in polychaeta at 26.2 ng g^{-1}. Acenaphthene was found in appreciable concentrations only in *Nucula sp.* (17.6 ng g^{-1}). Although the bivalves exposed to acenaphthene and acenaphthylene in the Hot sediments accumulated to higher levels than the New York Bight Apex field-collected organisms, the differences were variable and inconsistent.

a.



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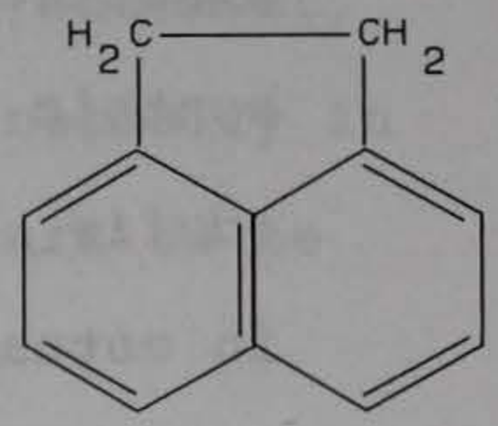


c.

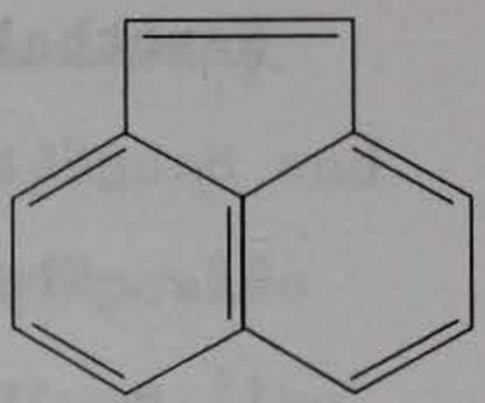


Figure 18. PAH structures:
a. naphthalene,
b. acenaphthene,
c. acenaphthylene

Phenanthrene, anthracene, and fluorene

Structures of the three PAH are shown in Figure 19. Phenanthrene and anthracene are three-ring planar PAH isomers but have differences in their physicochemical properties that affect bioavailability. The water

solubility of phenanthrene is 17-20 times greater than that of anthracene, and the log K_{oc} of phenanthrene is 4.08, whereas the log K_{oc} of anthracene is 4.20 (Karickhoff 1981). These characteristics make phenanthrene relatively more bioavailable to aquatic biota than is anthracene. Both bivalves bioaccumulated phenanthrene, but only traces of anthracene, from the Reference sediment. Mussels bioaccumulated both PAH compounds to approximately three-fold higher levels from suspended than from bedded Hot sediment, and the clams bioaccumulated to essentially the same concentrations from both types of exposure. Twenty-eight day tissue residues of phenanthrene in *M. nasuta* were 634 and 610 ng g^{-1} phenanthrene, and 237 and 216 ng g^{-1} anthracene, respectively, in the bedded and suspended Hot sediment exposures. Mussels bioaccumulated 49.5 and 138 ng g^{-1} phenanthrene and 13.8 and 42.5 ng g^{-1} anthracene in the bedded and suspended exposures.

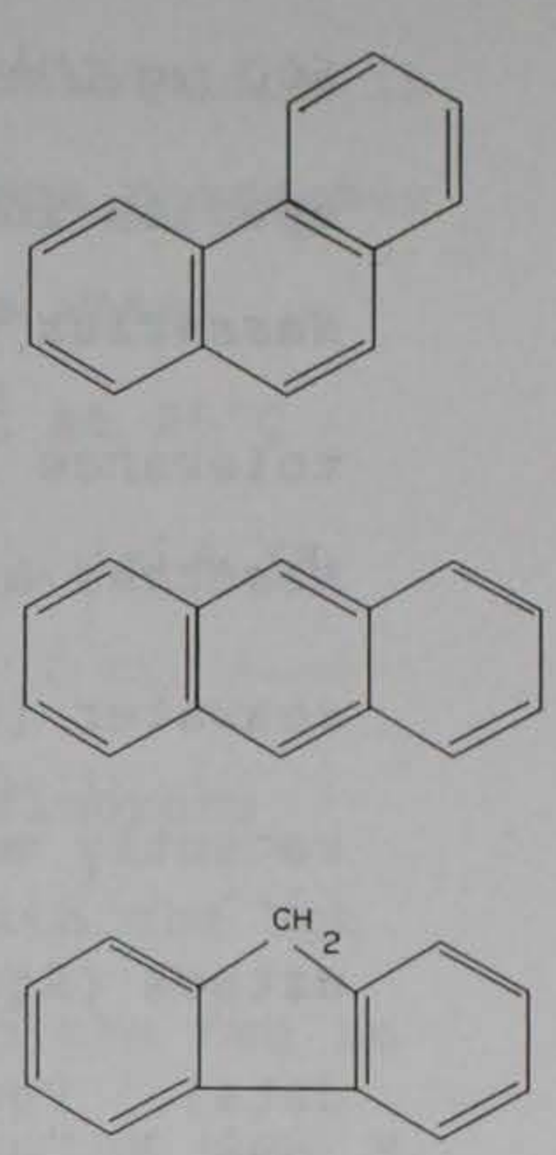


Figure 19. PAH structures:
a. phenanthrene
b. anthracene
c. fluorene

Phenanthrene is not carcinogenic or mutagenic and does not have high acute toxicity to most aquatic organisms. The

LC_{50}s for saltwater organisms were reported to range 21.9 to 600 $\mu g\ L^{-1}$ (Hanson et al. 1991a). The only molluscan species included in that survey was the marine snail, *Nassarius obsoletus*, with an $LC_{50} > 245\ \mu g\ L^{-1}$. The median tolerance limit (TLm) for phenanthrene to the polychaete *Neanthes arenaceodentata* was reported to be 600 $\mu g\ L^{-1}$ in seawater (Rossi and Neff 1978). The LC_{50} in sediment was recently measured at 660 mg Kg^{-1} to the amphipod, *Hyallela azteca* (Aquatic Contaminants Team, USAE WES, unpublished data). Fewer toxicity data have been reported for anthracene. However, anthracene is also reported to be noncarcinogenic and nonmutagenic in the Ames Test (reported in Verschueren 1983). No acute toxicity data was found for anthracene, but the 24-hr no effect level to trout was reported at 5 mg L^{-1}.

In the field studies at the New York Bight Apex site, phenanthrene was found in all taxa at concentrations ranging from 8.18 (*Mercenaria sp.*) to 90.5 ng g^{-1} (*Nucula sp.*). Anthracene was found in four of the seven taxa collected at concentrations from 1.04 (Polychaeta) to 23.9 ng g^{-1} wet weight (*Nucula sp.*).

Fluorene (Figure 19) is a three-ring, nearly coplanar unsaturated PAH. This compound bears an isosteric relationship to the nucleus of the polyhalogenated coplanar

hydrocarbons that include PCDDs, PCDFs, and coplanar PCBs. However, lacking chlorine or bromine-atom substitution in the lateral positions of the molecule, fluorene possesses none of the toxicity of those compounds. The water solubility of fluorene is given as 1.9 mg L^{-1} at 25°C (reported in Verschueren 1983) and the log K_{ow} is 4.18 (Hansch and Leo 1979).

Considering the 330-fold difference in fluorene concentration in the Reference as compared with the Hot sediment, and the relative similarity between the two in terms of organic carbon content, it is noteworthy that *M. nasuta* bioaccumulated to only an approximately eight-fold greater fluorene concentration from the Hot sediment than from the Reference. *Macoma nasuta* may affect its uptake of bioaccumulating chemicals through a mechanism of particle-size selection in feeding (Boese et al. in press 1994). Concentrations of fluorene in *M. edulis* reached approximately the same levels, ranging 2.00-6.77 ng g^{-1}, with higher uptake in the suspended sediment exposures.

Few field data are reported for fluorene body burdens in bivalves. Concentrations of fluorene in the estuarine filter-feeding clam, *Rangia cuneata*, transplanted to the vicinity of a creosote spill for up to four weeks were reported ranging 5-63 ng g^{-1} wet weight (DeLeon et al. 1988). In the seven taxa collected at the New York Bight

Apex field site, fluorene was quantitated only in *Nucula sp.* at a concentration of 16.8 ng g^{-1} wet weight.

Fluoranthene and pyrene

Both compounds are fully aromatic four-ring hydrocarbons. Although structurally dissimilar (Figure 20), the two compounds have similar water solubilities and log K_{ow}s. The solubility of fluoranthene is given as 0.265 mg L^{-1} at 25°C, and pyrene is reported to be 0.16 mg L^{-1} at 26°C (reported in Verschueren 1983). The log K_{ow}s for fluoranthene, biphenyl, and pyrene are summarized in Hanson et al. (1991b) with a single value, 5.155, reported for fluoranthene. Karickhoff (1981) reported a log K_{ow} = 5.18 for pyrene, and the log K_{oc} for that compound was measured at 4.83. These data indicate a similar bioavailability can be expected for the two compounds to aquatic organisms.

Clams exposed to bedded and suspended Hot sediment bioaccumulated fluoranthene to very high levels: 1785 and 1871 ng g^{-1}, respectively. Mussels exposed to the Hot sediments also bioaccumulated fluoranthene to high levels, particularly from the suspended sediment. Low concentrations (8.92-13.0 ng g^{-1}) were bioaccumulated by the two bivalves from Reference sediment. Mussels bioaccumulated low levels of pyrene from both bedded and suspended Reference sediments. Pyrene concentrations in clams exposed to the same sediments were not significantly

elevated after 28-day exposures. However, clams exposed to both bedded and suspended Hot sediments bioaccumulated high concentrations of pyrene (1750 and 1958 ng g^{-1}), respectively, as did mussels exposed to suspended Hot sediment (1415 ng g^{-1}). The Reference sediments produced negligible pyrene bioaccumulation in the bivalves, whereas the Hot sediment exposures resulted in tissue concentrations on the order of 30-fold greater than those reported in the Reference exposures, similar to the pyrene concentration differences in the two sediments.

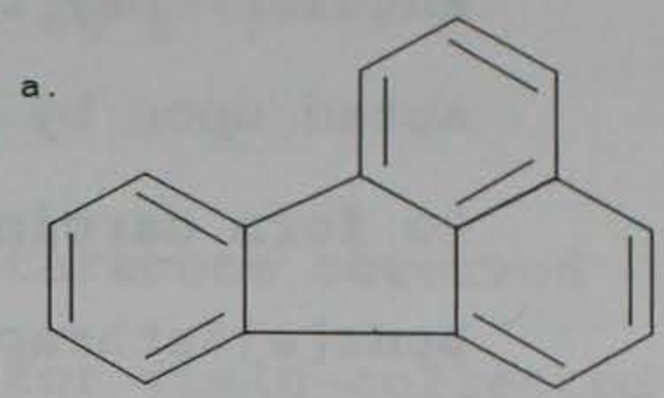


Figure 20. PAH structures:

a. fluoranthene
b. pyrene

Fluoranthene and pyrene were detected in all taxa collected at the New York Bight Apex field site. Concentrations were far below those analyzed in bivalves exposed to the Hot sediment, ranging 0.45 ng g^{-1} (*Cerebratulus lacteus*) to 113 ng g^{-1} (*Nucula sp.*) for fluoranthene, and 3.98 ng g^{-1} (*C. lacteus*) to 101 ng g^{-1} (*Nucula sp.*) for pyrene.

Benz[a]anthracene and chrysene

These two four-ring PAH compounds (Figure 21) are isomeric with pyrene, but unlike that compound possess the

angular "Bay region" that can be acted upon by CYPIA1 monooxygenases to form carcinogenic diol epoxides. Benz[a]anthracene and chrysene are mutagenic in the Ames Test and are considered weak carcinogens to humans (Williams and Weisburger 1986). The log K_{ow}s of chrysene and benz[a]anthracene are 5.70 and 5.91, respectively (cited in Mackay et al. 1992). The water solubility reported for chrysene is 0.006 mg L^{-1} at 25°C.

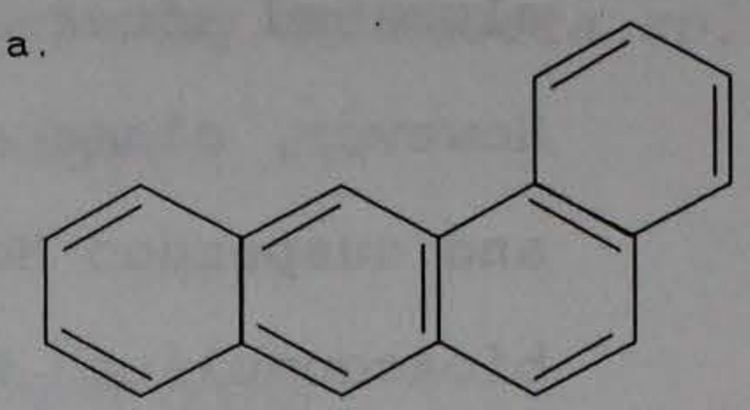


Figure 21. PAH structures

a. benz[a]anthracene
b. chrysene

A trace level of benz[a]anthracene (2.03 ng g^{-1}) was bioaccumulated by mussels from the suspended Reference sediment, and a much higher amount (623 ng g^{-1}) was taken up from the suspended Hot sediment. Clams bioaccumulated similar concentrations of benz[a]anthracene (452 and 492 ng g^{-1}) from suspended and bedded Hot sediment. Chrysene was bioaccumulated to high levels by both organisms from the Hot sediment. Statistically significant chrysene bioaccumulation in mussels occurred only in those organisms exposed to the suspended Hot sediment and was 925 ng g^{-1} at the end of the exposure period. Clams bioaccumulated chrysene from both bedded and suspended Hot sediment

exposures to similar levels: 630 and 697 ng g^{-1}, respectively.

The uptake of chrysene and benz[a]anthracene observed here far exceeds concentrations reported for field-collected organisms in TABLE III. Although the two PAH compounds were quantitated in nearly all organisms analyzed, the concentration range for benz[a]anthracene was only 0.38 (*C. lacteus*) to 52.8 ng g^{-1} (*Nucula sp.*), and for chrysene, 2.55 (*C. lacteus*) to 60.3 ng g^{-1} wet weight (*Nucula sp.*).

Benzo[a]pyrene, dibenz[a,h]anthracene, and benzo[b+k]fluoranthene

All the members of this group of five-ring PAH compounds are potent human carcinogens. Benzo[a]pyrene and dibenz[a,h]anthracene are isomers, as are benzo[b]- and benzo[k]fluoranthene (Figure 22). The addition of a benzyl ring to each of these compounds as compared with their four-ring homologues confers greater hydrophobicity (suggested log K_{ow}s, 6.04-6.20, Mackay et al. 1992) and/or forms the angular Bay region configuration necessary for bioactivation of the compounds to carcinogenic metabolites (Williams and Weisburger 1986). Benzo[a]pyrene and benzo[b+k]fluoranthene were bioaccumulated from Reference sediments by *M. nasuta* to similar levels, ranging 4.48-11.8 ng g^{-1} wet weight with highest concentrations reached in the suspended sediment exposures. Both the clams and the mussels bioaccumulated

the two compounds to levels of several hundred ng g^{-1} from bedded and suspended Hot sediment, with the highest concentration reached being 1278 ng g^{-1} benzo[b+k]-fluoranthene in *M. edulis* exposed to suspended sediment. These levels are several times greater than concentrations observed in the field study organisms. Field-collected polychaetes and mollusks at the New York Bight Apex site ranged in concentrations of benzo[a]pyrene and benzo[b+k]-fluoranthene from < 10 ng g^{-1} in *Nephtys sp.*, *C. lacteus*, and *Mercenaria sp.* to 50 to 90 ng g^{-1} for *Nucula sp.*, with miscellaneous mollusks and polychaetes intermediate at ≈ 10-33 ng g^{-1} wet weight.

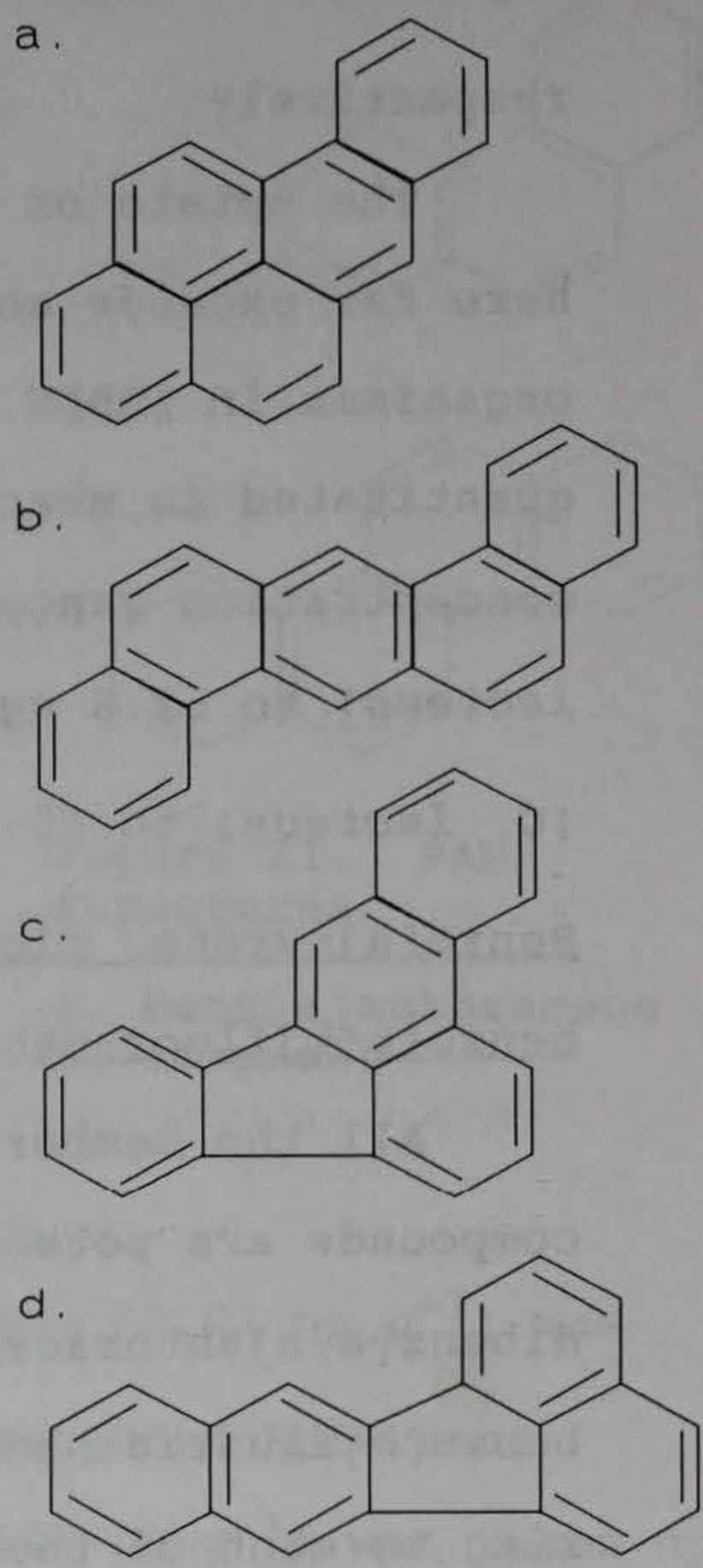


Figure 22. PAH structures:

a. benzo[a]pyrene
b. dibenz[a,h]anthracene
c. benzo[b]fluoranthene
d. benzo[k]fluoranthene

The concentrations of dibenz[a,h]anthracene bioaccumulated from the Hot bedded and suspended sediment by mussels (12.3 and 16.7 ng g^{-1}) and clams (25.0 and 27.0 ng g^{-1}) were comparable to dibenz[a,h]anthracene concentrations

in field-collected *Nucula sp.* (20.3 ng g^{-1}) and did not greatly exceed concentrations in other taxa at the New York Bight Apex field site (0.17-3.48 ng g^{-1}).

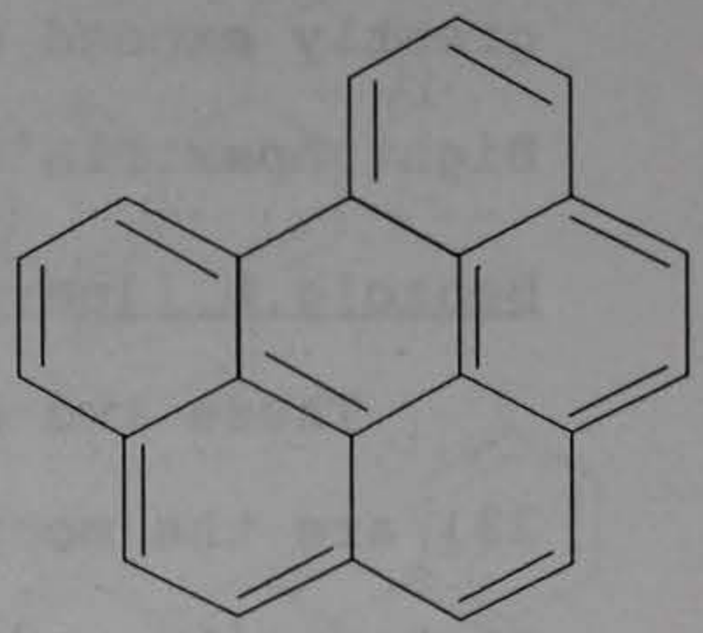
Benzo[g,h,i]perylene and indeno[1,2,3-cd]pyrene

These two six-ring unsubstituted PAH compounds (Figure 23) are the most hydrophobic and the least studied in terms of toxicity and bioaccumulation. Both compounds were bioaccumulated by *M. nasuta* from bedded and suspended sediment of both the Reference and Hot exposures. Bioaccumulation levels were very similar for the two compounds in all like exposures. Reference sediment-exposed clams bioaccumulated 4.15-5.9 ng g^{-1} benzo[g,h,i]perylene or indeno[1,2,3-cd]pyrene. Those exposed to Hot sediments bioaccumulated 71.9 and 85.2 ng g^{-1} Indeno[1,2,3-cd]pyrene, and 100 and 117 ng/g benzo[g,h,i]perylene from bedded and hot sediments, respectively. Mussels bioaccumulated only from Hot sediments with highest concentrations being reached in the suspended sediment exposures. Mussels bioaccumulated 172 ng g^{-1} benzo[g,h,i]perylene, and 136 ng g^{-1} indeno[1,2,3-cd]pyrene. With the exception of *Nucula sp.* at 43.8-163 ng g^{-1}, concentrations of the two PAH compounds bioaccumulated from the Reference sediment were in the range observed for field-collected organisms at the New York Bight Apex site (≤ 13.4 ng g^{-1} wet weight).

PAH toxicity to aquatic and marine invertebrates

Sediment-associated fluoranthene toxicity to amphipods has been shown to be inversely related to the organic carbon content of sediments, and an interstitial water 10-day LC_{50} = 23.8 μg L^{-1} was reported for *Rhepoxynius abronius* (Swartz et al. 1990). Acute toxicities of fluoranthene for saltwater organisms have been reported ranging from 1.6 μg L^{-1} for embryonic mysid shrimp, *Mysidopsis bahia*, to > 560,000 μg L^{-1} for the sheepshead minnow, *Cyrinodon variegatus*. A bivalve mollusk, the coot clam, *Mulinia lateralis* was reported to have a fluoranthene LC_{50} of 10,710 μg L^{-1} in seawater (Hanson et al. 1991b). The time at which the LC_{50}s were measured was not given. Fewer toxicity data are available for pyrene.

a.



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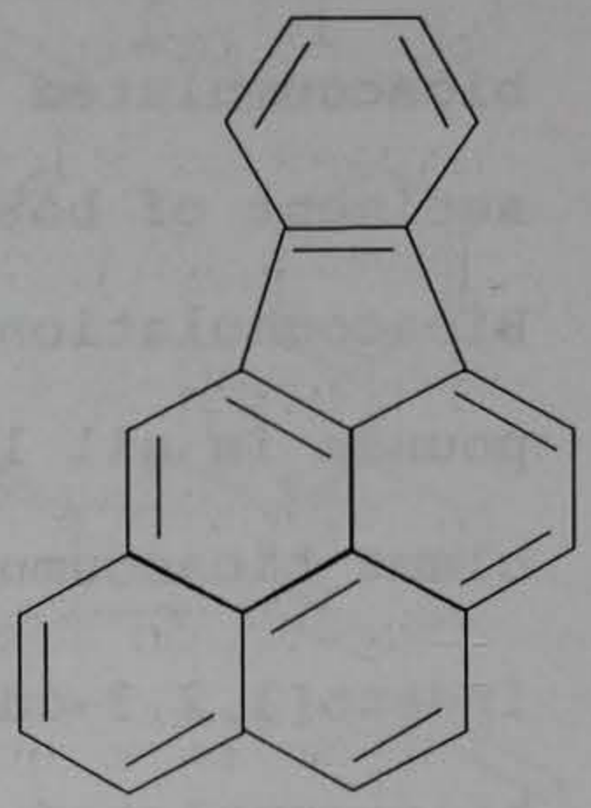


Figure 23. PAH structures:

a. benzo[g,h,i]perylene
b. indeno[1,2,3-cd]-pyrene

Toxicities caused by PAHs in bivalves have been related to their effects on lysosomal membranes. Derangement of the lysosomal structure and stability and associated loss of

lysosomal enzyme function in the digestive tract has been observed in *M. edulis* exposed to phenanthrene and anthracene (Moore and Farrar 1985). A critical concentration threshold was observed for lysosomal effects in mussels exposed to phenanthrene, but with anthracene the effects were linear with concentration in tissues. Major dysfunction occurred with both PAHs at tissue concentrations above 20 $\mu g\ g^{-1}$ in the laboratory studies. These concentrations are far in excess of anthracene and phenanthrene concentrations bioaccumulated in the two bivalves from either Reference or Hot sediments in the present study. However, it is possible to speculate based on the type of toxicity observed, that effects on membranes of PAH compounds are more likely due to the molar concentration of total PAH compounds and less dependent on concentration of a specific compound. This inference is drawn from the results of numerous studies in membrane fluidity that have demonstrated the ability of lipophilic chemicals to disrupt the function of membrane-bound enzymes (Gennis 1989). Tables XVI-XIX show that the total concentration of the 15 PAH compounds analyzed in clams and mussels exposed to the Hot sediment ranges 2,244-7,289 ng g^{-1}. In a population of bivalves chronically exposed to the Hot sediment, it appears possible that lysosomal toxicities would result. However, the Reference sediment does not approach these concentrations in 28-day exposures.

An obvious question is, "What is the relevance of tissue residues measured in 28-day exposures to body burdens in organisms receiving life-time exposures in the field?" Would exposure of the bivalves to the Reference sediments be expected to result in potentially toxic concentrations, given a sufficiently long exposure?

KINETICS CONSIDERATIONS

Although the relative concentrations of neutral organic compounds at equilibrium in organic phases are not correlated with K_{ow} (Broman et al. 1990, Ferraro et al. 1990, Lake et al. 1990) partition coefficients can be used to describe the kinetics of bioaccumulation. Equations were derived by Spacie and Hamelink (1982), based on the work of others, correlating uptake and elimination rate constants for neutral organic chemicals in fish with their K_{ow}s. The relationship of hydrophobicity to time to steady state (t_{ss}) for organic compounds based on numerous studies of single chemical bioconcentration in fish describes an inverted parabola (Spacie and Hamelink 1982). Bioconcentration increases from log $K_{ow} < 3$ to a peak at log $K_{ow} \approx 6$, and then decreases to the limits of bioconcentrating or bioaccumulating compounds, log $K_{ow} \approx 10$. Connell and Hawker (1988) described the phenomenon using a polynomial expression enabling t_{ss} to be estimated from the log K_{ow} of a compound. Clarke and McFarland (1991) applied the

equation derived by Connell and Hawker (1988) to the estimation of steady-state concentrations, C_{ss}, from tissue residue data obtained in 28-day exposures (or other exposure periods less than steady state).

The single time-point estimation of C_{ss} is based on the simple first-order one-compartment model most commonly used in bioaccumulation and bioconcentration kinetics studies (Branson et al. 1975, Connell 1990, Könemann and van Leeuwen 1980, Mackay 1982, Spacie and Hamelink 1982), i.e.:

$$dC_t/dt = k_1C_w - k_2C_t \qquad (9)$$

where C_t is concentration in organism tissue, C_w is concentration in water (or sediment, if that is considered the source), and k_1 and k_2 are the uptake and elimination rate constants, respectively. When integrated for constant exposure, the model takes the form:

$$C_t = ((k_1C_w)/k_2)(1 - e^{-k_2t}) \qquad (10)$$

As time t approaches infinity, the term e^{-k_2t} approaches zero and C_t becomes C_{ss}:

$$C_{ss} = (k_1C_w)/k_2 \qquad (11)$$

which relates rates of uptake and elimination, and the exposure concentration to the concentration at steady state. Equation 10 can be used to derive:

$$t = -\ln(f_{ss})/k_2 \qquad (12)$$

which enables calculation of the time required to reach a fraction of steady state, f_{ss}.

The polynomial relationship calculated by Connell and Hawker (1988) is:

$$\log t_{ss} = 6.9 \times 10^{-3}(\log K_{ow})^4 - 1.85 \times 10^{-1}(\log K_{ow})^3 + 1.65(\log K_{ow})^2 - 5.34(\log K_{ow}) + 5.93 \quad (13)$$

The time required to reach steady-state bioaccumulation or bioconcentration (t_{ss}) of a neutral organic compound is estimated from its log K_{ow} using equation 13. From the t_{ss}, an estimation of k_2 can be made by rearranging equation 12 and substituting t_{ss} for t, as:

$$k_2 = -\ln(1 - f_{ss})/t_{ss} \quad (14)$$

A value of f_{ss} near 1 (e.g., 0.99) is used because the approach to steady state is asymptotic. Equation 12 can be rearranged to solve for f_{ss}:

$$f_{ss} = 1 - e^{-k_2 t} \quad (15)$$

and the fraction of steady state reached by 28 days of exposure can be estimated using t = 28 and using the k_2 estimated from equation 14. Finally, the concentration projected to result at steady state based on the log K_{ow} and the estimate of f_{ss} from equation 15 is:

$$C_{ss} = C_t/f_{ss} \quad (16)$$

where C_t is the measured concentration in an organisms tissues after 28 days of exposure, and C_{ss} is the predicted concentration at steady state based on that single time-point measurement.

The log K_{ow}s of the 15 PAH analytes measured in sediments and biota of the field and laboratory studies are listed in TABLE XXIV (compiled in Mackay et al. 1992). These were used as described to estimate f_{ss} (28-days), t_{ss}, and estimate C_{ss} for the PAHs bioaccumulated by mussels from suspended Reference sediments (TABLE XXV). The data set was selected because it contained the highest concentrations bioaccumulated in the Reference sediment exposures.

Steady state bioaccumulation is estimated to have been reached within the 28-day exposure period for six PAH compounds: naphthalene, acenaphthene, acenaphthylene, fluorene, phenanthrene, and anthracene. For pyrene and fluoranthene to reach steady state is estimated to take more than two months. The remaining compounds appear to require from more than four months, to nearly one year, to bioaccumulate to steady state. At 28-days, the most slowly bioaccumulating PAHs, benzo[g,h,i]perylene and indeno[1,2,3-cd]pyrene appear to achieve one-third their steady state concentrations. The effect on total concentration of the 15 analytes appears to be to increase the sum from 120 ng g^{-1} only to approximately 136 ng g^{-1}, a 14-percent increase.

TABLE XXIV

MOLECULAR WEIGHTS AND SELECTED OR RECOMMENDED OCTANOL/WATER PARTITION COEFFICIENTS OF PAH COMPOUNDS

Compound	m.w.	log K_{ow}[a]
Naphthalene	128.16	3.45
Acenaphthene	156.23	3.92
Acenaphthylene	154.21	4.08
Fluorene	166.22	4.18
Phenanthrene	178.22	4.50
Anthracene	178.22	4.63
Pyrene	202.25	5.18
Fluoranthene	202.25	5.22
Chrysene	228.28	5.70
Benz[a]anthracene	228.28	5.91
Benzo[a]pyrene	252.30	6.04
Dibenz[a,h]anthracene	252.30	6.20
Benzo[b or k]fluoranthene	252.30	6.20
Benzo[g,h,i]perylene	276.34	7.10
Indeno[1,2,3-cd]pyrene	276.34	7.10

[a]Compiled in Mackay et al. 1992.

Actual bioaccumulation will be influenced by numerous variables, both dependent on characteristics of the exposure medium and on physiological and metabolic characteristics of the organism. Estimations from physicochemical properties and simple models require interpreting with a sense of the uncertainty inherent in them. The estimations made for the PAH compounds in the Reference sediments indicate that 28-day exposures are probably sufficient to describe body burdens that could result given longer exposures in the field and suggest that the concentrations reached would not

TABLE XXV

ESTIMATES BASED ON OCTANOL/WATER PARTITION COEFFICIENTS CONTRASTED WITH MEASURED 28-DAY TISSUE CONCENTRATIONS OF PAH COMPOUNDS IN MUSSELS, *M. EDULIS*, EXPOSED TO SUSPENDED REFERENCE SEDIMENTS

Compound	t_{ss}[a] (days)	f_{ss} (28-day)[b]	C_t (28-day)[c]	Est. C_{ss}[d]
Naphthalene	3.36	1.00	44.1	44.1
Acenaphthene	6.88	1.00	1.65	1.65
Acenaphthylene	9.05	1.00	0.13	0.13
Fluorene	10.80	1.00	3.27	3.27
Phenanthrene	19.22	1.00	30.60	30.60
Anthracene	24.31	1.00	2.50	2.50
Pyrene	62.61	0.87	16.00	18.33
Fluoranthene	66.74	0.86	10.32	12.07
Chrysene	132.87	0.62	4.51	7.26
Benz[a]anthracene	170.27	0.53	2.03	3.82
Benzo[a]pyrene	194.88	0.48	0.58	1.20
Dibenz[a,h]anthracene	225.42	0.44	0.0	0.00
Benzo[b+k]fluoranthene	225.42	0.44	3.64	8.35
Benzo[g,h,i]perylene	325.86	0.33	0.47	1.44
Indeno[1,2,3-cd]pyrene	325.86	0.33	0.31	0.95
Total of 15 PAH			120.11	135.71

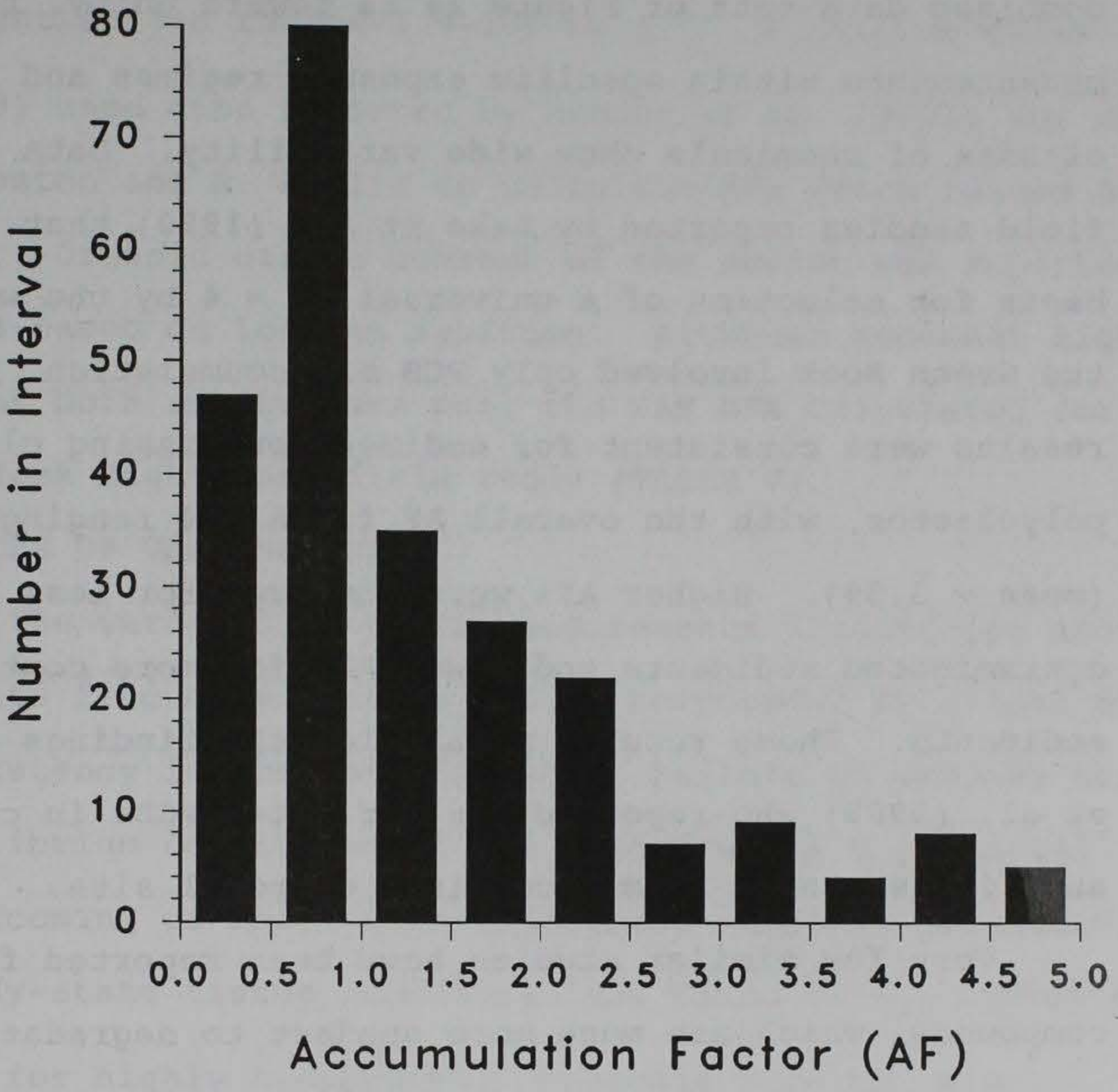
[a]Time to steady state (equation 13).
[b]Fraction of steady state concentration at 28 days (equation 15).
[c]Measured tissue concentration after 28-days exposure.
[d]Estimated steady-state tissue concentration (equation 16).

be of a magnitude likely to cause toxicities in bivalve mollusks.

EMPIRICAL AFS VS THE UNIVERSAL AF

Since the early 1980's a number of laboratory and field investigations have been performed in which the AF (or BSAF) was measured empirically for a variety of organisms and chemicals. Polychlorinated biphenyls have been the subject of most published field and laboratory investigations in which AFs were measured, and chlorinated pesticides, chlorobenzenes, PCDDs, PCDFs, chlorinated phenols and butadienes, and PAHs account for the rest (Parkerton et al. 1993). When data from the literature were analyzed after expression on a common basis for lipid extraction, i.e., chloroform/methanol (Randall et al. 1991) a skewed distribution of AFs resulted (Figure 24) with a mean AF = 1.009 (n = 250, SE = 0.059), and median AF = 0.650 (25%ile = 0.390, 75%ile = 1.370).

Conversion of pf (1.73) from a hexane/acetone lipid basis to an equivalent chloroform/methanol lipid basis (Randall et al. 1991) results in pf = 0.927. Neither mean nor median AF appears to differ greatly from the idealized pf = 0.927. When the data set was tested for normality (Kolmogorov-Smirnov test), it was found to be significantly non-normally distributed ($P < 0.001$) and a nonparametric test (Wilcoxon signed rank test) was used to compare the pf with the median AF. There was no significant difference



between the two (P = 0.2702).

Figure 24. Frequency distribution of AFs reported in the literature[a]. All lipid data not expressed on a chloroform/methanol extraction basis converted using Randall et al. 1991.

[a]Ankley et al. 1992; Boese et al. [1994]; Brannon et al. 1989, 1991; Clarke et al. 1988; Ferraro et al. 1990, 1991; Foster et al. 1987; Lake et al. 1987, 1990; McElroy and Means 1988; McFarland et al. 1994, Pruell et al. 1990, 1993; Young et al. 1991.

Although the central tendency of AF values in the combined data sets of Figure 24 is toward unity, the AF measurements within specific exposure regimes and for classes of chemicals show wide variability. Data of the field studies reported by Lake et al. (1990) that formed the basis for selection of a universal AF = 4 by the writers of the Green Book involved only PCB bioaccumulation. The results were consistent for sediment-processing clams and polychaetes, with the overall AF for A1254 ranging 1.14-7.27 (mean = 3.39). Higher AFs were observed for less contaminated sediments and lower AFs for more contaminated sediments. These results paralleled the findings of Clarke. et al. (1988) who reported AFs for total PCBs in crawfish and finfish taken from a confined disposal site.

Very few similar studies have been reported for PAH compounds, which are much more subject to degradative processes than are PCBs or other chlorinated hydrocarbons. However, two studies have been reported involving PAH AFs calculated for the bivalve mollusks, *M. nasuta*, and *M. edulis*. In both studies PAH AFs were intermediate between the AFs for PCBs reported by Lake et al. (1990) and the AFs for PAHs calculated from data of the New York Bight Apex field study. Ferraro et al. (1990) reported AFs for pyrene, chrysene, benzo[a]pyrene, benz[a]anthracene, and benzo[b+k]fluoranthene ranging 0.05-1.02. *Macoma nasuta* were exposed to bedded sediment in the laboratory for 28

days, and the sediments ranged 0.86-7.37% TOC. Sediment PAH concentrations ranged 1.4-186 ng g^{-1}. Parkerton et al. (1993) used data reported by Broman et al. (1990) for PAHs in seston and *M. edulis* to calculate AFs which ranged 0.02-0.46. Organic carbon content of the seston was reported as 26.8% based on loss on ignition. Although somewhat higher, AFs of both studies are near the PAH AFs calculated for the New York Bight Apex field study (TABLE V).

SOURCES OF VARIABILITY

The variability in AF measurements illustrated above results from many factors and is compounded by a lack of consistency in the methods used. Failure to achieve near equilibrium conditions in the exposures is a potential shortcoming of short-term exposures. Kinetic projections of steady-state tissue concentrations based on K_{ow} indicate that for highly hydrophobic chemicals such as, e.g., benzo[g,h,i]perylene, only about one-third of the steady-state concentration will be reached in a 28-day exposure (TABLE XXV). However, in our laboratory bioaccumulation studies the projected steady-state concentration of the total of 15 PAH analytes in the Reference sediment was increased by only about 14-percent over the concentration reached after 28-days of exposure.

A greater source of variability in both laboratory and field studies may result from the use of organisms that are not in intimate and continuous contact with sediments. For

example, mussels exposed to bedded Hot sediment only through the water column did not experience the same bioavailability as did mussels that were able to filter the Hot sediment when it was suspended (TABLE XI). Similarly, AFs calculated for lake trout, which normally have little or no contact with sediments, or for ducks that forage on sediment but range widely, are highly variable and do not reflect equilibrium relationships (Parkerton et al. 1993).

Although long-term laboratory exposures represent a partial solution to the non-equilibrium problem, they have their own difficulties. During long-term studies, sediments may be depleted of the most bioavailable fraction of a chemical, thereby reducing exposure, and resulting in lower tissue concentrations than would be expected based on sediment chemistry. Nutrient quality and amount may decline and sublethal toxicity may occur, affecting the health of organisms and causing loss of lipids. Metabolic degradation of chemicals that are taken up may occur, also reducing bioaccumulation levels. Induction of metabolizing enzymes caused by the chemical under investigation or by other chemicals in the sediment may exacerbate this effect. Growth during the exposure period can dilute tissue concentrations causing reduced apparent bioaccumulation. Spawning and other seasonal changes also affect bioaccumulation (Lee et al. 1989).

Additionally, the chemical must be fully bioavailable, or nearly so: if desorption of the chemical from sediment binding sites is very slow, an exposed organism may never reach equilibrium bioaccumulation. In the results reported here, we have seen that bioaccumulation was not a linear function of organic carbon-normalized PAH concentration in the Reference and Hot sediments. This observation probably reflects the interaction of several of the variables cited, and it also can be speculated that PAHs analyzed in the Hot sediment included a relatively large sequestered fraction, not readily capable of desorption. Karickhoff (1980) modeled a biphasic approach to equilibrium for sorption of pyrene, phenanthrene, and naphthalene on suspended sediment particles consisting of a "rapid component" with time constants ranging 4 to > 30 hr^{-1}, and a slow component ranging 0.056 to 1.5 hr^{-1}. The "slow component" was visualized as being bound within a deeper organic carbon matrix of sediment particles and inaccessible to the mass of surrounding water. Release of organic chemicals from the deeper matrix would require diffusion to interfacial sites. Such a process would be impeded by a succession of sorptive steps which would slow the diffusion from the interior to the surface of the particle similarly to the elution of an analyte from a chromatography column. For higher molecular weight PAHs, the result could be a much lesser

bioavailability to sediment-exposed organisms than would be expected based on sediment chemistry.

All of these processes influence the kinetics of phase distribution of a chemical between sediment and organism, i.e., alterations in the rates of chemical transfer between phases can alter the value assigned to the endpoint, AF, which is intended to express a thermodynamic relationship.

Other sources of variability not related to length of exposure include loss of linearity in the contaminant/TOC relationship at low TOC concentrations where mineral surfaces and other factors play an increased role in sorption dynamics. In sediment-processing organisms the selective ingestion of particles according to size or nutritive value may result in a different dose to the animal than is represented by normalizing on the basis of TOC in the whole sediment (Boese et al. in press 1994). In addition, types of organic carbon present in the sediments, and differences in lipid composition, life-style of the organism and presence and levels of other contaminants in the sediments may affect the AF (McElroy and Means 1988). Procedural differences in analysis of TOC and lipid can also influence the result.

Lipid normalization of concentration data using the same extraction procedure for both laboratory and field studies enabled AFs calculated for the seven taxa representing the infaunal and epifaunal benthic assembly at

the New York Bight Apex field site to be used to predict PAH bioaccumulation in unrelated organisms from the West Coast. Also, organic carbon content of the sediments used to generate the AFs, and those for which bioaccumulation potential was predicted differed by only about a factor of two. These two factors very likely played a large role in the accuracy of the predictions made in the present study. A further refinement of the TBP model would consider the influences of different lipid extraction procedures and of a wider range of organic carbon contents. The influence of a range of concentrations of the bioaccumulating chemicals would also be included as a term in a refined TBP model.

SUMMARY AND CONCLUSION

In the investigations reported here, the predictive capability of a screening procedure currently used in regulating open-water disposal of dredged sediments was compared with that of a modification to the procedure. The procedure applies equilibrium partitioning principles to the estimation of bioaccumulation potential from sediment chemistry. As presently recommended in the "Green Book," the testing manual for Public Law regulating dredged material disposal (U.S. Environmental Protection Agency/U.S. Army Corps of Engineers 1991), a universal accumulation factor, AF, based on results of bioaccumulation studies involving PCB compounds, is used to represent all neutral organic chemicals.

Field-generated AFs for specific PAH compounds were used in the modified screening procedure. Benthic organisms representing seven taxa were collected by grab sampler at a site on the continental shelf of the New York Bight Apex. Concentrations of PAH compounds were measured in the tissues of the organisms and in the sediment in which they were collected. The concentration data were normalized on organism lipid content and sediment organic carbon content, for calculation of the individual PAH AFs.

In a separate laboratory study, two bivalve mollusks not included in the taxa of the field study collection were exposed for 28 days to PAH contaminated sediments collected

from the San Francisco Bay system. Concentrations of PAH compounds were measured in the SF Bay sediments, as were concentrations in tissues of the experimental organisms before and after the exposures.

Using the concentration data of the SF Bay sediments, estimations of the sediment-associated PAH bioaccumulation potential were made in two ways: (1) using the universal AF = 4 as currently recommended in the Green Book, and (2) using the AFs for specific PAH compounds generated in the field study at the New York Bight Apex site.

Bioaccumulation potential estimations made by each procedure were compared with actual bioaccumulation measured in the laboratory study. For the PAH compounds, estimations using the field-generated AFs were predictive of the actual measured bioaccumulation. However, estimations made using the Green Book recommended AF = 4 were highly inaccurate and grossly overestimated bioaccumulation.

The investigations reported in this dissertation demonstrate the inaccuracy of bioaccumulation potential estimations from sediment chemistry made using a relationship based primarily on PCBs when applied to bioaccumulation of PAH compounds. However, the application of relationships observed for specific PAH compounds in a field situation were shown to result in accurate predictions of bioaccumulation potential measured in the laboratory.

I conclude from these results that, in the case of sediment-associated PAH compounds, regulatory practice can be improved substantially by replacing the universal AF currently used in Green Book sediment bioaccumulation potential estimations with AFs derived for specific compounds from data of appropriately designed field bioaccumulation studies. By inference, the potential for improvement of bioaccumulation potential estimations involving other classes of organic chemicals appears to exist.

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Equilibrium partitioning of neutral organic chemicals between the organic carbon fraction of bedded sediments and the lipids of resident organisms provides the theoretical basis for one of the most popular approaches to the development of sediment quality criteria (SQC) by the U.S. Environmental Protection Agency (EPA). The proposed equilibrium partitioning-based SQC seek to relate estimated doses of sediment-associated chemicals to toxicity in exposed biota. Criteria documents for several polynuclear aromatic hydrocarbon (PAH) compounds, endrin, and dieldrin have been released by the EPA for public review, and may soon be promulgated. A procedure recommended in the Implementation Manual (the "Green Book") for public law regulating ocean disposal of dredged sediments (Section 103, Public Law 92-532, Marine Protection, Research, and Sanctuaries Act, the "Ocean Dumping Act") has used equilibrium partitioning-based estimations to screen sediments for bioaccumulation potential for several years. The screening test, termed "theoretical bioaccumulation potential," TBP, is also included in the draft manual for inland waters to implement dredged material testing requirements of the Clean Water Act. TBP employs an accumulation factor (AF), defined as the ratio at equilibrium of the organic carbon-normalized concentration of a neutral organic chemical in a sediment and the lipid-normalized concentration of the

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chemical in an exposed organism. The Green Book currently recommends using a universal AF = 4 for all neutral chemicals, the rationale being that this value is suitably protective of all neutral chemicals, provided certain caveats are recognized.

This study compared the predictive capability of PAH AFs derived from field data with that of the universal AF = 4 in making TBP estimations. Predicted bioaccumulations using the two methods was compared with PAH tissue concentrations measured in laboratory exposures of clams (*Macoma nasuta*) and mussels (*Mytilus edulis*).

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