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Environmental Effects of Dredging
Technical Notes

Literature Review for Residue-Effects Relationships with Hydrocarbon Contaminants in Marine Organisms

Purpose

The purpose of this literature review was to identify potential residue-effects relationships involving hydrocarbon contaminants which are described in the scientific literature. That information will be used to develop guidance for interpreting the results of bioaccumulation experiments conducted in the regulatory evaluation of dredged material.

Background

Work Unit 31771, "Environmental Interpretation of Consequences from Bioaccumulation," of the Long-Term Effects of Dredging Operations (LEDO) Program is designed to generate interpretive guidance for evaluating data produced by Corps field offices or their permit applicants. This guidance results from identifying residue-effects relationships through laboratory experiments and literature reviews. Previous investigations have focused on two classes of environmental contaminants--heavy metals and chlorinated hydrocarbons. The current effort examines residue-effects relationships with hydrocarbon contaminants by a literature survey.

Hydrocarbons are an extremely complex class of environmental contaminants consisting of aliphatic, cyclic, aromatic, and heterocyclic compounds (Blumer 1976). Most of the toxicity of petroleum hydrocarbons to aquatic organisms is due to the aromatic fraction (Anderson and others 1974, Rice, Short, and Karinen 1977, Neff and others 1976). Because aromatic hydrocarbons are composed of one or more aromatic rings they are called polycyclic aromatic hydrocarbons (PAHs).

PAHs are ubiquitous environmental contaminants (Neff 1979). They are most often associated with the accidental release of petroleum, but may also originate

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from pyrolytic and biogenic sources. Origin notwithstanding, PAHs tend to partition into sediments due to their hydrophobic nature. Consequently, when sediments are scheduled for dredging, the bioavailability of PAHs to aquatic organisms may need to be evaluated.

In 1987, the US Army Engineer Waterways Experiment Station (WES) conducted a workshop in which experts recommended that 15 of the 16 priority pollutant PAHs should be analyzed during the regulatory evaluation of dredged material (Clarke and Gibson 1987). Naphthalene, a diaromatic hydrocarbon, was omitted from the list because the workshop participants felt it was too volatile for routine chemical analysis and did not persist in sediments. It was also felt that if high levels of naphthalene were present in sediment, its effects would be manifested as mortality in acute toxicity bioassays.

Subsequent to that workshop, a tiered testing protocol for dredged material containing hydrocarbon contaminants was developed (Jarvis and Clarke 1990). One of the tiers (Tier III) includes bioaccumulation testing using deposit-feeding organisms that have little or no metabolic capability for PAHs. For example, most fish and aquatic invertebrates rapidly metabolize PAHs while marine bivalves have little or no such capability (Lee, Sauerheber, and Benson 1972, Varanasi 1989). However, the interpretive guidance to assess the results of these sediment bioaccumulation tests is currently lacking.

Additional Information

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Approach

Published literature reporting the effects of PAHs on marine organisms was reviewed. Only investigations which examined organismic endpoints in bivalve molluscs such as growth, reproduction, behavior, and metabolism were included. Bivalve molluscs were emphasized because they have little or no biotransformation capability and they are the species of choice for assessing the bioaccumulation potential of PAHs in dredged material (Jarvis and Clarke 1990). Organismic sublethal effects are desirable endpoints for the regulatory evaluation of dredged material for reasons previously discussed (Dillon 1984). Anderson (1977) also concluded that growth, reproduction, and behavior may be the most sensitive and meaningful biological measures when the effects of petroleum hydrocarbons on aquatic organisms are being evaluated.

More than 30 technical journals and 10 data base literature search services (for example, Biosis, Pollution Abstracts, and National Technical Information Service) were used in this review. Over 100 publications were individually reviewed. For each paper included in this review, the following information was recorded: test

species, exposure conditions, hydrocarbon tissue concentration, and corresponding biological effects.

Analysis

Publications which contained both hydrocarbon residue and biological effects information for marine bivalve molluscs are shown in Table 1. All investigations evaluated the effects of crude or refined oil exposed via water or sediment. Laboratory investigations slightly outnumbered field studies and all the latter focused exclusively on exposure via oiled sediment. There were no acute exposures. Laboratory exposures ranged from 28 days to 16 months. The duration of field studies ranged from 38 days to 6 years. The longer term exposures were part of monitoring studies conducted after the accidental release of petroleum. All investigations were limited to only four species of bivalve molluscs--the filter-feeding blue mussel, *Mytilus edulis;* the soft-shell clam, *Mya arenaria;* and the deposit-feeding bivalves, *Macoma* sp. and *Protothaca staminea.*

Direct and indirect measures of growth were the most popular biological endpoints. One such measure, Scope For Growth (SFG), has been studied extensively in the mussel *Mytilus edulis* (Bayne 1985). This endpoint is an instantaneous measure of growth based on the amount of calories consumed less the amount required for maintenance and lost via excretion. If there are excess calories after calculating SFG, the mussel is said to have a positive SFG. Negative SFG values are generally indicative of stressful conditions and have been strongly associated with diminished reproduction in this mussel.

Another measure of growth, Condition Index (CI), evaluates the amount of bivalve tissue relative to its shell size or volume (Lawrence and Scott 1982). The advantage of measuring growth via this endpoint is that differences among molluscs in their shell size are normalized. If the CI is reduced, then the amount of tissue relative to its shell size or volume has decreased. One underlying assumption is that a change in tissue mass occurs more rapidly than shell size. This is a reasonable assumption to make.

Tissue concentrations in most investigations are expressed as aromatic hydrocarbons--total, diaromatic, or triaromatics. The range of concentrations spans four orders of magnitude. Three investigations reported residues as total aliphatics, while two reported total hydrocarbons. To more clearly evaluate potential residue-effects relationships, those aromatic hydrocarbon tissue concentrations in Table 1 which are associated with adverse biological effects were ranked in descending order (Table 2). The highest tissue concentrations (about 200-300 µg/g) are reported as total aromatics, while the lowest concentrations (about 0.01-1.0 µg/g) are found when residues are expressed as di- or triaromatic hydrocarbons. Remaining tissue residues are in the double-digit µg/g range.

A wide variety of analytical methods were used to analyze for hydrocarbons in bivalve tissue (Table 3). Most investigators used gas chromatography (GC) or high performance liquid chromatography (HPLC). With appropriate extraction

techniques, either can be used to quantify both aromatic and aliphatic hydrocarbons. Three studies used ultraviolet absorption or fluorometry which are specific to aromatic hydrocarbons. Total hydrocarbons were analyzed in two papers using infrared spectrometry and gravimetric analysis.

Conclusions

Only a small proportion (about 10 percent of publications reviewed contained information on both the biological effects of hydrocarbons and the corresponding tissue residues in marine bivalves. Similar results were reported earlier for other environmental contaminants and aquatic biota (Dillon 1984). This small data base greatly restricts the ability to generate quantitative guidance on hydrocarbon residue-effects relationships. In addition to a small data base, variations in analytical methods reduce the effectiveness of any potential guidance.

Despite these difficulties, some general qualitative trends are apparent from the data reviewed. For example, biological effects are associated with relatively high tissue concentrations (about 200-300 μg/g) when those data are expressed as total aromatics. Lower body burdens are observed if aromatic hydrocarbons groups (for example, di- and triaromatics) are reported individually (about 1-100 μg/g) or together (about 0.01-1.0 μg/g). Moore and others (1987), in reviewing numerous papers on the effects of petroleum on field-exposed mussels, reported a similar range of effects-related tissue concentrations (1-100 μg/g) for di- and triaromatic hydrocarbons. Anderson (1977, 1979) reviewed the effects of petroleum hydrocarbons on fish, crustaceans, and polychaetes and found adverse effects at tissue concentrations of 0.2-0.6 μg/g total naphthalenes or 0.2-10.0 μg/g total aromatics.

Are these data sufficient to provide interpretive guidance for the regulatory evaluation of dredged material? Unfortunately the answer is no. The data base is too small and does not provide any specifics regarding the 15 individual PAHs on the priority pollutant list.

Two approaches for developing the needed guidance on PAHs are possible. One approach is the generation of site-specific guidance based on tissue concentrations in organisms collected in and around the disposal site environs. This so-called matrix approach assumes a local policy of "no further degradation" and that the environmental status quo is acceptable. The advantage to this approach is that numerical guidance can be generated with relative ease. There are three primary disadvantages. The field-collected organisms must be the same or closely related to the sediment bioassay test species. The toxicological significance of the bioassay results is unknown. For example, how does one interpret results where only one of the 15 priority pollutant PAHs is accumulated or 3 out of 15 or 8 of 15 are accumulated? Finally, there is no allowance for ecological interpretation. All comparisons are statistical. Tissue concentrations slightly but significantly above matrix values are treated the same as grossly elevated residues but different from concentrations slightly below but significantly different from matrix values.

The second approach is the ecotoxicological approach, which requires more effort than the matrix approach, but provides additional interpretive latitude. Here the toxicological significance of the priority pollutant PAHs is determined individually and as a group. Ideally, the model organism is the same as or closely related to the sediment bioassay test species. Next, guidance on the ecological significance of bioaccumulation is developed by generating residue-effects relationships for the individual PAHs. With these data, the ecological and toxicological importance of PAH bioaccumulation can be interpreted in a technically sound manner.

Naphthalene was not included on the experts' list of PAHs. This omission may warrant further consideration because many of the residue-effects papers reported diaromatic (naphthalenic) hydrocarbons concentrations, di- and triaromatic hydrocarbons are the major constituents in mussels from oil-contaminated environments (Boehm and others 1982, Farrington and others 1982), and di- and triaromatic hydrocarbons contribute most to the toxicity of petroleum (Neff and others 1976, Rice, Short, and Karinen 1977, Anderson and others 1974).

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Table 1

Literature Reporting Tissue Hydrocarbon Concentrations in Marine Bivalves and Corresponding Biological Effects

Reference	Contaminant	Organism	Exposure Time	Exposure Concentration	Biological Effect*	Tissue Concentration
1**	Prudhoe Bay crude oil	*Protothaca staminea*	54 days (field)	850-1,237 µg/g (sediment)	CI reduced	0.184 diaromatics 0.104 triaromatics 0.428 aliphatics
2	Prudhoe Bay crude oil	*Macoma inquinata*	55 days (laboratory)	616-1,233 µg/g (sediment)	CI reduced	1.15-5.21 total naphthalenes 0.14-0.42 aliphatics
			38 days (field) (exp. 2)	364-1,144 µg/g (sediment)	CI reduced	0.01-0.07 total naphthalenes 0.42-0.46 aliphatics
3	Prudhoe Bay crude oil	*Macoma balthica*	180 days (laboratory)	30-3,000 µg/L (water)	CI reduced; growth reduced	81-350 total aromatics 68-240 aliphatics

(Continued)

* Text of Footnote

** Numbered references are given at the end of the table; full bibliographic information is given in the References section.

NOTE: Tissue concentrations are given in micrograms per gram wet weight, unless otherwise noted.

Table 1 (Continued)

Reference	Contaminant	Organism	Exposure Time	Exposure Concentration	Biological Effect	Tissue Concentration
4	Bunker C	*Mya arenaria*	6 years after spill (field)	3,800 µg/g (sediment)	Growth reduced	267 total aromatics
5	Bunker C	*Mya arenaria*	6 years after spill (field)	5,115 µg/g (sediment)	Growth reduced	157 total aromatics
6	Diesel oil	*Mytilus edulis*	8 months (laboratory) (exp. 1)	28-125 µg/L (water)	SFG reduced	2.9-68.5 di- + triaromatics
			8 months (laboratory) (exp. 2)	28-125 µg/L (water)	SFG reduced; growth reduced	0.71-128.1 di- + triaromatics
7	Diesel oil	*Mytilus edulis*	8 months (laboratory)	30-130 µg/L (water)	SFG reduced; feeding reduced	21-24 di- + triaromatics
8	North Sea crude oil	*Mytilus edulis*	28 days (laboratory)	36 µg/L (water & food)	SFG reduced; food absorption reduced	21.8-78.3 aromatics (digestive gland)
					Oxygen consumption elevated	8.8-16.2 aromatics (remaining tissue)
3	Prudhoe Bay crude oil	*Macoma balthica*	180 days (laboratory)	30 µg/L (water)	Oxygen consumption unaffected	81 total aromatics 68 aliphatics

(Continued)

Table 1 (Continued)

Reference	Contaminant	Organism	Exposure Time	Exposure Concentration	Biological Effect	Tissue Concentration
				300-3,000 µg/L (water)	Oxygen consumption reduced	130-150 total aromatics 160-240 aliphatics
9	No. 2 fuel oil	*Mya arenaria*	28 days (laboratory)	4,500 µg/L (water)	Oxygen consumption elevated	20-30 total hydrocarbons
10	No. 2 fuel oil	*Mya arenaria*	28 days (laboratory)	43.7-60.7 mg/L (water)	Oxygen consumption unaffected	60-145 total hydrocarbons
11	No. 6 fuel oil	*Mya arenaria*	1 year after spill (field)	11.8 mg/g (sediment)	Oxygen consumption elevated	661 total hydrocarbons
12	Diesel oil	*Mytilus edulis*	4-16 months (laboratory)	29-123 µg/L (water)	Gamete number reduced	14.7-25.4 diaromatics 3.4-7.4 triaromatics
8	North Sea crude oil	*Mytilus edulis*	140 days (laboratory)	30 µg/L (water)	Gamete production unaffected	152 aromatics (digestive gland) 22.9 aromatics (remaining tissue)
2	Prudhoe Bay crude oil	*Macoma inquinata*	55 days (field) (exp. 1)	88-1,233 µg/g (sediment)	Abnormal surfacing in sediments	0.01-0.05 total naphthalenes

(Continued)

Table 1 (Concluded)

Reference	Contaminant	Organism	Exposure Time	Exposure Concentration	Biological Effect	Tissue Concentration
						0.06-0.14 aliphatics
3	Prudhoe Bay crude oil	*Macoma balthica*	180 days (laboratory)	30-3,000 µg/L (water)	Burrowing rate (unaffected)	81-350 total aromatics
						68-240 aliphatics

1. Augenfeld and others 1980
2. Roesijadi and Anderson 1979
3. Stekoll, Clement, and Shaw 1980, Clement, Stekoll, and Shaw 1980
4. Gilfillan and Vandermeulen 1978
5. Thomas 1978
6. Widdows, Donkin, and Evans 1987
7. Widdows, Donkin, and Evans 1985
8. Widdows and others 1982
9. Stainken 1976
10. Stainken 1978
11. Gilfillan and others 1976
12. Livingstone and others 1985

Table 2

Effects-Level Tissue Concentrations (µg/g wet weight) of Hydrocarbons in Marine Bivalves*

Reference	Aromatics		Aliphatics	Total Hydrocarbons
3	81-350	total	68-240	
4	270	total		
5	160	total		
8	22-78	total (dig. gland)		
	8.8-16	total (rem. tissue)		
6	0.71-130	di- + triaromatics		
6	2.9-68	di- + triaromatics		
7	21-24	di- + triaromatics		
12	15-25	diaromatics		
	3.4-7.4	triaromatics		
2	1.2-5.2	diaromatics	0.14-0.42	
1	0.18	diaromatics	0.43	
	0.10	triaromatics		
2	0.01-0.07	diaromatics	0.42-0.46	
2	0.01-0.05	diaromatics	0.06-0.14	
11				660
9				20-30

* Residues are taken from Table 1 and rounded to two significant figures; see Table 1 for references.

Table 3

Methods Used to Analyze Bivalve Tissues for Hydrocarbon Content*

Analytical Method	Reference
Gas chromatography/glass capillary column	1, 2, 3
Gas chromatography/packed column	10
Ultraviolet absorbance	4
Ultraviolet absorbance with GC/MS confirmation	8
High performance liquid chromatography	6, 7, 12
Fluorometry	5
Infrared spectrometry	9
Gravimetric	11

* See Table 1 for references.

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