FRESHWATER BIOLOGICAL ASSOCIATION

PETROLEUM HYDROCARBONS IN FRESH WATERS

A Preliminary Desk Study and Bibliography

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by

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1. INTRODUCTION

To understand the effects of petroleum hydrocarbons in fresh waters, we require information on their toxicity, biodegradability and concentration. To obtain this information a survey of the literature was carried out, supplemented by a selective search on hydrocarbons in the marine environment, for purposes of comparison. Our aim has been to determine the major inputs of these hydrocarbons, their accumulation, effects and fate in fresh waters, as revealed by the published literature. This information is summarized in Tables 3 - 10 below.

The search was confined to the period 1965 - 1978, since there was little significant study of hydrocarbons in fresh waters before then, and the general field of hydrocarbon microbiology prior to 1965 is covered by several excellent reviews.* Since 1965 a number of large oil-spills have resulted in a rapid growth of the literature, and recent work has produced a better understanding of the biochemical mechanisms of oil decomposition.

The main bibliographic sources used in the search were as follows:

Aquatic Biology Abstracts, Vols 2-3, 1970-1971, continued as
Aquatic Sciences and Fisheries Abstracts, Vols 1-8, 1971-1978.
Biological Abstracts, Vols 46-66, 1965-1978.
Bioresearch Index, Vols 10-14, 1974-1978.
International Biodeterioration Bulletin Reference Index Supplement, Vols 5-8, 1969-1972, continued as
Biodeterioration Research Titles, Vols 9-14, 1973-1978.
Pollution Abstracts, Vols 1-9, 1970-1978.
Water Pollution Abstracts, Vols 38-46, 1965-1973, continued as
WRC Information, Vols 1-5, 1974-1978.
A bibliography on marine and estuarine oil pollution, Suppl. 2, by D.S. Moulder & A. Varley (1978). Plymouth, Marine Biological Association of the U.K.

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* e.g. Davies, J.B. (1967). *Petroleum microbiology*. Amsterdam, Elsevier. 604pp.
Van der Linden, A.C. & Thijsse, G.J.E. (1965). The mechanisms of microbial oxidation of petroleum hydrocarbons. *Adv. Enzymol.* 27, 496-546.

The terms used for searching included: hydrocarbons; petroleum; petrochemicals; diesel fuel; oil; gasoline; benz-pyrene; bacteria; micro-organisms; pollution.

About 800 references were retrieved, of which 390 were examined in some depth; these latter form the bibliography which follows*. The final selection was based on relevance, originality, scientific quality and language - we were unable to obtain translations of Russian and other papers in the time available.

Of the papers retrieved, 82% were found in one bibliographic source only. A breakdown of information by environment is shown in Table 1, and by geographical origin in Table 2; these tables should be interpreted in accordance with the selective nature of the search as described above, and the somewhat random inclusion of certain material. It is unlikely, for example, that North America has produced no papers on carcinogenesis, and the results merely reflect that only a few papers, mainly Russian in origin, were chosen for inclusion. Figures for the relatively small British contributions to work on microbial decomposition and environmental analysis (Table 2, B,C) may be considered as more reliable since the sample sizes were much larger.

* In a few cases only the abstract was seen; in consequence, bibliographic details may be incomplete.

Table 1 Breakdown of references by environment

Reference classification	*Environment (%)*			
	Freshwater	*Marine*	*General aquatic*	*Laboratory*
A. Effects, Uptake, Accumulation (155 papers)	28*	29	10	33
B. Microbial Utilization, Degradation (91 papers)	33	36	4	27
C. General Occurrence, Properties (36 papers)	76	3	15	6
E. Causes, Sources (37 papers)	89	8	3	0
Total Papers	41	23	7.5 (+ 3.5 in soil)	25

* figures are percentages, for each environment, of the papers retrieved under each reference class. Final row is environmental breakdown for all the papers in these classes, including those on soil.

Table 2 Breakdown of references by geographical origin

Reference classification	*Origin*				
	British	*European*[1]	*N.American*	*Other*	*Total*
A. Effects, Uptake, Accumulation (155 papers)	12*	11	72	5	40
B. Microbial Utilization, Degradation (91 papers)	7	16	73	4	23
C. General Occurrence, Properties (36 papers)	6	40	43	11	9
D. Methods (38 papers)	12	18	70	0	10
E. Causes, Sources (37 papers)	22	31	44	3	10
F. Treatment (15 papers)	6	47	47	0	4
G. Carcinogenesis (5 papers)	20	80	0	0	1
H. Soil (13 papers)	18	45	37	0	3
Total	11.5	21	63	4.5	100

* figures are percentages of the papers retrieved under each reference class. The final row is the geographical breakdown for all the papers abstracted in this study. The final column is the percentage breakdown of all papers abstracted by class.

[1] Including USSR

2. CLASSIFICATION OF THE REFERENCES

References are classified and the bibliography arranged in subject groups as follows. In each group, references are in alphabetic order of authors and are identified by the section letter and a serial number.

A. *Ecological and toxic effects, uptake, accumulation:* including toxicity tests, observed uptake in all organisms and accumulation down the food chain.

B. *Microbial utilization, degradation:* laboratory and environmental studies.

C. *General occurrence, properties:* including analyses of samples from natural and man-made environments.

D. *Methods, detection and analysis:* the most commonly used methods of analysis.

E. *Causes, sources:* details of the inputs of petroleum hydrocarbons to the environment.

F. *Treatment:* in the environment and in effluent plants.

G. *Carcinogenesis:* this section (along with F) was not within the remit of the contract but the most relevant references abstracted were retained.

H. *Soil:* not a complete survey, but selected references were included to obtain an understanding of microbial action on hydro-carbons before entry into water courses.

A. ECOLOGICAL AND TOXIC EFFECTS, UPTAKE, ACCUMULATION

This section is mainly concerned with the effects of petroleum hydrocarbons on organisms. Detailed reviews on this subject are to be found in A 56, 74, 91, 96, 99, 101, 104, 110, 111, 117, 141 and 150. The literature on the harmful effects on macrobiota is summarized in Table 3, and general measurements of tissue concentrations not necessarily associated with damage are shown in Table 4. Hydrocarbons may stimulate or inhibit microorganisms, depending on the concentration and the species involved. A summary of the papers dealing with these effects is found in Table 5, and the more general environmental studies, classified by the communities and organisms involved, are presented in Table 6.

Notes

a. Units of concentration are variable and their meaning is not always clear. The units used by the original author are presented here, with no attempt at interpretation. No attempt is made to differentiate studies on single hydrocarbons or petroleum mixtures.
b. A large proportion of the work has been devoted to higher animals, especially fish. The adverse effects noted include the occurrence of tainting. Work particularly devoted to the effect of outboard engine exhaust products includes A 58, 59 and 64.
c. Microorganisms may be stimulated by hydrocarbons and can play a part in their decomposition. Mutagenesis is amongst the harmful effects noted (A 113).
d. Much of the work on microorganisms has been done with pure cultures or pure hydrocarbons.
e. The range of toxic concentrations, in all cases, is large.
f. The possible excretion of polluting hydrocarbons by macrobiota requires further consideration (A 27, 133).

Table 3 Concentration ranges of hydrocarbons observed to affect biota adversely

Organisms	*Concentration range*		*References*
	In organism	*In environment*	A
Fish	0.7 - 94 ppm	0.03 - 3500 ppm	2,3,8,19,28,34, 35,40,41,68,82, 83,86,87,88,92, 93,94,114,115, 116,139
Decapods	0.14 - 1.6 ppm	0.95 - 1000 ppm	3,17,137
Amphipods	0.3 - 0.4 ppm	0.1 - 4.2 ppm	3,17,44
Polychaetes	0.3 - 20 ppm	-	3,120,121
Insects	-	14 - 59 ppm	12,13,14
Shellfish	10 ppm	0.05 - 17 ppm	15,24
Birds	12.5 - 50 ppm (oral dose)		29,30

Table 4 Concentration ranges of hydrocarbons observed in biota

Organisms	*Concentration range*	*References* A
Fish	0.1 - 1000 ppm	1,4,27,106,107
Shellfish	0.8 - 69 ppm	4,98,100
Amphipods	43 ppm	4
Ducks		72,73
Benthic invertebrates	0.02 - 210 $\mu g\ g^{-1}$	134,138
Algae	15 - 30 $\mu g\ g^{-1}$	140

Table 5 The effect of petroleum hydrocarbons on microorganisms

Organism and effect (see footnote)	Hydrocarbon concentration	References A
B+, Ac, Pc	0.1 - 1.0 ml ml^{-1}	5
A+	10 - 30 μg l^{-1}	7
A-	0.038 - 0.124 mg l^{-1}	9,16
A-	> 0.001 ppm	18
B-	< 2% saturation	22
B-	> 1.2 mg l^{-1}	23
A-	0.1 - 10 ml l^{-1}	31
A-	0.1 - 10%	33
Bo/-, Ao/-	-	36
A+/-	10^2 - 10^4 μg l^{-1}	39
B+	80 ppb	53
B-	> 300 ppb	53
B+	-	55
A-	33 - 1780 ppm	61,62
A-	3 - 500 ppm	63
A-	0.1 - 1.0 g l^{-1}	66
A+	< 5 ppm	69
A-	5 - 700 mg l^{-1}	70
B+	1000 ppm	90
A-	-	103
A+/-	5 - 500 μg l^{-1}	112
A-	10 - 100 mg l^{-1}	128-132,E3
B+, Fo	-	146
B-	-	147
A-	5 ppm	151-153

Abbreviations : First letter: A = algae, B = bacteria, P = protozoa, F = fungi

Second letter: + = stimulation, - = inhibition, +/- both effects depending on concentration, o = no change, c = change in species composition.

Table 6 General environmental studies

Environment	*Community - organisms*		*Effects*	*References* A
Stream	Benthos	algae	species change	10
	"	fauna	lethal	21
	"	algae	harmful	25
		macroinvertebrates	toxic	54
	"	"	lethal	81
	"	"	reduction in numbers	97
River	Benthos	macrofauna, bacteria	some toxicity,	84,85
	"	chironomids	effect species-dependent	118
Lake	Benthos	chironomids	lethal	11
	"	littoral	lethal	126,127
	"	invertebrates sublittoral invertebrates, algae	increase in numbers	126,127
Pond	Plankton	algae	decreased efficiency	26
	"	"	no effect	50
	Benthos	"	inhibition	50
	"	insects	lethal	60
	Plankton	algae, bacteria	stimulation	124
	Plankton	algae	inhibition	143
Marine	Plankton	algae	inhibition	80
	General flora and fauna		lethal	89
Saltmarsh	Higher plants		harmful	6, 20
	Bacteria		increase in numbers	42

B. MICROBIAL UTILIZATION, DEGRADATION

The papers abstracted in this section are classified in Table 7.

Notes

a. The relative importance of bacteria in aquatic decomposition processes is in sharp contrast to that observed in soil where the fungal contribution is greater.

b. Much of the degradation work is laboratory-based and reliable estimates of rates of decomposition in the field are relatively few.

c. There is mounting evidence of nutrient limitation of hydrocarbon decomposition in water (B 3, 6, 53, 85, 86).

d. Whereas the weight of evidence indicates preferential utilization of certain hydrocarbons, some studies (B 3, 59) have shown all fractions to be decomposed at similar rates. Growth on a single hydrocarbon is not a good indication of its utilization in a mixture (B 17).

e. There is evidence both of success (B 54, 66, 68) and failure (B 84, A 135) in attempts to obtain anaerobic decomposition of hydrocarbons in the presence of alternative electron transport acceptors.

f. Weathering may remove inhibitory substances from oil (B 2) but residual resinous tars may be more refractory than the volatile fraction (B 18).

g. It is of interest that the latest text in this subject area (B 13) contains no information on the effect of hydrocarbons on the microbiology of fresh waters.

Table 7 Classification of papers on microbial utilization, degradation Section B *

	Reviews	*Laboratory studies*	*Field studies*
Bacteria	21,22,34,35,36	2,4,7,15,18,25,27,44, 45,53,54,58,59,63,66, 68,70,71,72,76,79,80, 81,83,90,A144	3,9,32,33,49, 50,52,56,57, 60,69,73,74, 84,85,86,A125
Fungi	1	17,26,55	9,14,74
General	10,12,13,16,31, 38,40,41,46,48, 67,88	11,23,24,39,61,64,65, 78,82	8,29,30,37,42, 62,64,65

* Materials from other sections have letter prefix

C. GENERAL OCCURRENCE, PROPERTIES

A useful general review on allochthonous and autochthonous hydrocarbons in the aquatic environment is provided in C 3, and D 25 lists the hydrocarbons isolated from petroleum in recent analyses. A survey which concentrated on polynuclear aromatics (C 5) also included the results of toxicity tests on a range of petroleum hydrocarbons. Some of the data on observed concentration ranges are summarized in Table 8. Not all the references cited contain quantitative information. The potential carcinogenicity of polynuclear aromatic hydrocarbons (PAH) has undoubtedly contributed to the emphasis placed on this group. Baseline values for PAH and threshold odour concentrations of a number of compounds are published (E 4, C 19). The increase in concentration with proximity to centres of population suggests that combustion of fossil fuels may be the main source of PAH (C 12). The concentration ranges of the hydrocarbons in Table 8 overlap those shown to exert an adverse effect on macrobiota (Table 3) and microorganisms (Table 5).

Table 8 Some analyses of hydrocarbons in freshwaters

Hydrocarbon	*Concentration range*	*References* C
Aliphatic	1400 $\mu g\ g^{-1}$ *	33
Polynuclear aromatic	10^{-5} - 13 $\mu g\ l^{-1}$	1,7,10,18,28
Crude/fuel oil	0.02 - 1.42 $mg\ l^{-1}$	8,21,22,23,24
Petroleum/products	0.15 - 0.8 $mg\ l^{-1}$	16,25
Total hydrocarbons	50 - 1500 $\mu g\ g^{-1}$	6,9,26,29,32
	1 - 5.5 $\mu g\ l^{-1}$	13,14,20,A33

* analyses of sediments are calculated on a dry weight basis

D. METHODS, DETECTION AND ANALYSIS

Although not primarily concerned with methodology this desk study retrieved several papers concerned with analytical procedures. These are summarized in Table 9. Further details of methodology, particularly with regard to changes in composition due to microbial activity are to be found in references cited in Section B. Gas chromatographic procedures have been used extensively and these are coupled with analysis of mass spectra when detailed analysis is required.

Table 9 Methods used for analysis of petroleum hydrocarbons in the natural environment

Method	*References* D
Gravimetric, physico-chemical	6,7,11,14,16,20,27,31
Resin sorption	1,34
Paper chromatography	7,27
Column chromatography	1,9,15,23,35,38
Thin-layer chromatography	1,7,16,33,37
High pressure liquid chromatography	3,12,30
Gas chromatography	1,4,5,6,7,9,10,15,17,18,20,22, 24,27,29,32
Visible and U.V. spectroscopy	7,8
Infra-red spectroscopy	2,7,14,18,19,20,26,27,28
Fluorescence spectroscopy	2,3,7,13,27,35,38
Mass spectrometry	8,9,15

Table 10 Major sources of petroleum hydrocarbons in aquatic environments

Environment	*Assessment of pollution sources*[1]	*References*[2] E
Water supplies	Largely from oil pipelines	1
Subsurface/ground water	Waste dumping, settling tanks, pipelines	2,5,27,31
Lakes/Reservoirs	a) Lake Champlain: 38% run off, leaks, dumping; 6% shipping traffic; 56% outboard motor boats	A109
	b) Lake Washington: 70% river and stormwater damage; 11% bridge runoff; 10% dustfall; 3% rainfall; 6% streamflow; < 0.4% outboard motor boats	34,35
	c) 50% of pollution due to heating oil (gas oil) or diesel oil from road vehicles	D37
	d) Fuel oil from machinery, stormwater surface runoff	7
	e) Outboard motor exhaust	9,11
	f) Industry, navigation, underwater pipelines	22,23
General inland waters	a) 45% storage, pipelines; 5% tanker deliveries; 7% garages, boats etc; 8% road tanker accidents; 35% general incidents	29,C30
	b) Tipping, blockages, seepage, overfilling tanks, garages, accidents	25,A32
	c) Mainly heating and diesel fuel; sewage effluents and surface runoff may have adverse long term effects	13
	d) Storm water	14
	e) Outboard motors	15,19,28
	f) PAH, mainly from airborne combustion products	16
	g) Industry and distribution	20,26

Table 10 (continued)

Environment	*Assessment of pollution sources*[1]	*References*[2] E
Rivers	a) 30% domestic and industrial heating accidents during filling and storage; 10% pipelines, 10% waste oil disposal; 11% engines (including service stations, depots, airfields); 5% tanker accidents; 23% other traced sources; 11% untraced sources	23
	b) Run off	17
	c) Oil refinery and petrochemical wastes	24
	d) Municipal wastewater, secondary treatment plant	32
Effluents	Up to 20 mg l^{-1} petroleum hydrocarbons, oil and grease	12,C15,C34
Marine	On shore sources: 41% municipal wastes, non-refining industrial wastes, urban and river runoff	6,36,A56
Air	Municipal waste combustion	8

[1] Many studies were non-quantitative and the method of assessment must be obtained from the reference

[2] References from sections other than E have letter prefix

E. CAUSES, SOURCES

The assessment of sources of pollution has been largely non-quantitative. In some instances the relative importance of various sources has been estimated and these values are included in Table 10. In spite of the qualitative nature of much of the information the following points were considered to be of interest:

a. The relative importance of sources of lake pollution will vary enormously with lake usage (cf. results from Lake Champlain and Lake Washington in Table 10).
b. Many of the surveys have been concerned with catastrophic incidents. Chronic low-level pollution may be overlooked.
c. Although there have been several papers on the effects of outboard motors, many have provided only qualitative information. There appears to be some agreement that two-stroke lubricating oil is a major source of pollution (this would be reduced if four-stroke engines were used). Fish tainting has been observed at a boating density equivalent to consumptions of 8 volumes of outboard motor fuel per million volumes of lake water (E 18).

F. TREATMENT

Of the 15 references retrieved under this subject heading the majority were concerned with cleaning up catastrophic spills by collection, burning off or emulsification (F 2, 3, 7 and 9). The detention period of conventional sewage plants is too short for effective treatment (F 10) and alternative procedures must be adopted. Some success has been achieved with graphite and other sorbent filter processes (F 4, 5 and 11) although chlorination can modify polycyclic aromatic hydrocarbons (F 6). Treatment of contaminated groundwater is a difficult process (F 14, 15) and has not met with much success (F 8).

G. CARCINOGENESIS

All the references in this subject category were concerned with polycyclic aromatic hydrocarbons.

H. SOIL

Addition of oil stimulates the soil microflora (H 1, 4, 6, 7, 9) and there is some evidence of selection of fungi (H 5, 10), possibly related to the lead content of the pollutants. Although significant quantities of hydrocarbons may permeate soil strata to pollute water courses (particularly groundwater), decomposition in the soil may be inhibited in the absence of adequate aeration and mineral nutrition (H 2, 8, 12, 13).

3. CONCLUSIONS

1. Much of the published work on pollution by petroleum hydrocarbons has been concerned with catastrophic spillages of crude and fuel oil, particularly into the sea.

2. The results of such catastrophic events are often readily measured, but there is a need for a greater understanding of the effects of chronic low-level pollution.

3. Quantitative information on the relative importance of sources of petroleum hydrocarbons in fresh water is meagre, particularly in relation to water usage. Outboard motors, sewage effluent and surface runoff should receive particular attention.

4. There are relatively few reliable estimates of microbial decomposition of petroleum products in the aquatic environment. The decomposition of mixtures of hydrocarbons (rather than individual substrates) should be studied in greater detail.

5. Observed concentration ranges of petroleum hydrocarbons in water bodies overlap those shown to exert an adverse effect on macrobiota and microorganisms. The concentration and excretion of potentially toxic substances along freshwater food chains requires further investigation.

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Section D. Methods

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6. AUTHOR INDEX

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