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IMO /FAO/UNESCO/WMO/WHO/IAEA/UN/UNEP
JOINT GROUP OF EXPERTS ON THE SCIENTIFIC ASPECTS
OF MARINE POLLUTION
- GESAMP -

# **REPORTS AND STUDIES**

No. 40

1989

LONG-TERM CONSEQUENCES
OF LOW-LEVEL MARINE CONTAMINATION

AN ANALYTICAL APPROACH





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IMO/FAO/Unesco/WMO/WHO/IAEA/UN/UNEP
Joint Group of Experts on the Scientific Aspects of Marine Pollution
(GESAMP)

LONG-TERM CONSEQUENCES OF LOW-LEVEL MARINE CONTAMINATION: An Analytical Approach

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### DEFINITION OF MARINE POLLUTION BY GESAMP

"Pollution means the introduction by man, directly or indirectly, of substances or energy into the marine environment (including estuaries) which results in such deleterious effects as harm to living resources, hazards to human health, hindrance to marine activities including fishing, impairment of quality for use of sea water and reduction of amenities".

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### PREPARATION OF THIS STUDY

This document is based on the work of the GESAMP Working Group on Long-term Ecological Consequences of Low-level Contamination of the Marine Environment. The Working Group met from 13 to 17 July 1987 in Plymouth, U.K. The meeting was attended by B.L. Bayne, M. Bernhard, G.W. Brian, J.M. Colebrook, V. Dethlefsen, L. Eldredge (Rapporteur), V. Hongskul, G.D. Howells (Chairman), A. Kapauan, A.D. McIntyre H. Naeve (Technical Secretary), T.H. Pearson and A.J. Southward.

The present document has been prepared by a small core group, consisting of D. Calamari, J. Gray, G.D. Howells (Chairman), H. Naeve (Technical Secretary) and P.G. Wells. Meetings were held in Rome, Italy, 6-8 June 1988, Cambridge, U.K., 8-9 August 1988, and Rome, Italy, 4-6 October 1989.

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### Reports and Studies GESAMP

The following reports and studies have been published so far. They are available from any of the organizations sponsoring GESAMP.

- 1. Report of the seventh session, London, 24-30 April 1975. (1975). Rep.Stud. GESAMP, (1):pag.var. Available also in French, Spanish and Russian
- 2. Review of harmful substances. (1976). Rep.Stud.GESAMP, (2):80 p.
- 3. Scientific criteria for the selection of sites for dumping of wastes into the sea. (1975). Rep.Stud.GESAMP, (3):21 p. Available also in French, Spanish and Russian
- 4. Report of the eighth session, Rome, 21-27 April 1976. (1976). Rep.Stud.GESAMP, (4): pag.var. Available also in French and Russian
- Principles for developing coastal water quality criteria. (1976). Rep.Stud. GESAMP, (5):23 p.
- 6. Impact of oil on the marine environment. (1977). Rep.Stud.GESAMP, (6):250 p.
- 7. Scientific aspects of pollution arising from the exploration and exploitation of the sea-bed. (1977). Rep.Stud.GESAMP, (7):37 p.
- 8. Report of the ninth session, New York, 7-11 March 1977. (1977). Rep.Stud. GESAMP, (8):33 p. Available also in French and Russian
- 9. Report of the tenth session, Paris, 29 May 2 June 1978. (1978). Rep.Stud. GESAMP, (9):pag.var. Available also in French, Spanish and Russian
- 10. Report of the eleventh session, Dubrovnik, 25-29 February 1980. (1980). Rep.Stud.GESAMP, (10):pag.var. Available also in French and Spanish
- 11. Marine Pollution implications of coastal area development. (1980). Rep.Stud. GESAMP, (11):114 p.
- 12. Monitoring biological variables related to marine pollution. (1980). Rep.Stud. GESAMP, (12):22 p. Available also in Russian
- 13. Interchange of pollutants between the atmosphere and the oceans. (1980). Rep.Stud.GESAMP, (13):55 p.
- Report of the twelfth session, Geneva, 22-29 October 1981. (1981). Rep.Stud. GESAMP, (14):pag.var. Available also in French and Russian
- 15. The review of the health of the oceans. (1982). Rep.Stud.GESAMP, (15):108 p.
- 16. Scientific criteria for the selection of waste disposal sites at sea. (1982). Rep.Stud.GESAMP, (16):60 p.

- 17. The evaluation of the hazards of harmful substances carried by ships. (1982). Rep.Stud. GESAMP, (17):pag.var.
- 18. Report of the thirteenth session, Geneva, 28 February 4 March 1983. (1983). Rep.Stud. GESAMP, (18):50 p. Available also in French and Spanish
- 19. An oceanographic model for the dispersion of wastes disposed of in the deep sea. (1983). Rep.Stud.GESAMP, (19):182 p.
- 20. Marine pollution implications of ocean energy development (1984). Rep.Stud. GESAMP, (20):44 p.
- 21. Report of the fourteenth session, Vienna, 26-30 March 1984. (1984). Rep.Stud. GESAMP, (21):42 p. Available also in French, Spanish and Russian
- 22. Review of potentially harmful substances. Cadmium, lead and tin. (1985). Rep.Stud. GESAMP, (22):114 p.
- 23. Interchange of pollutants between the atmosphere and the oceans (part II). (1985). Rep.Stud. GESAMP, (23):55 p.
- 24. Thermal discharges in the marine environment. (1984). Rep.Stud.GESAMP, (24):44 p.
- 25. Report of the fifteenth session, New York, 25-29 March 1985. (1985). Rep.Stud. GESAMP, (25):49 p. Available also in French, Spanish and Russian
- 26. Atmospheric transport of contaminants into the Mediterranean region. (1985). Rep.Stud.GESAMP, (26):53 p.
- 27. Report of the sixteenth session, London, 17-21 March 1986. (1986). Rep.Stud. GESAMP, (27):72 p. Available also in French, Spanish and Russian
- 28. Review of potentially harmful substances. Arsenic, mercury and selenium. (in press). Rep.Stud.GESAMP, (28)
- Review of potentially harmful substances. Organosilicon compounds (Silanes and Siloxanes). (1986). Printed in limited number only by IMO, but published also as UNEP Reg. Seas Rep. Stud., (78):24 p.
- 30. Environmental Capacity. An approach to marine pollution prevention. (1986). Rep.Stud.GESAMP, (30):49 p.
- 31. Report of the seventeenth session, Rome, 30 March 3 April 1987. (1987). Rep. Stud.GESAMP, (31):36 p. Available also in French, Spanish and Russian
- 32. Land-sea boundary flux of contaminants: contributions from rivers. (1987). Rep.Stud.GESAMP, (32):172 p.
- 33. Report on the eighteenth session, Paris, 11-15 April 1988. (1988). Rep.Stud. GESAMP, (33):56 p. Available also in French, Spanish and Russian

- 34. Review of potentially harmful substances. Nutrients. (in press). Rep.Stud. GESAMP, (34)
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- 36. Pollutant modification of atmospheric and oceanic processes and climate: some aspects of the problem. (in press). Rep.Stud.GESAMP, (36)
- 37. Report of the nineteenth session, Athens, 8-12 May 1989. (1989). Rep.Stud. GESAMP, (37):47 p. Available also in French, Spanish and Russian
- 38. Atmospheric input of trace species to the world ocean. (in press). Rep.Stud. GESAMP, (38)
- 39. The state of the marine environment. (in press). Rep.Stud.GESAMP, (39)
- 40. Long-term consequences of low-level marine contamination: An analytical approach. (1989). Rep.Stud.GESAMP, (40):14 p.

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## LONG-TERM CONSEQUENCES OF LOW-LEVEL MARINE CONTAMINATION: An Analytical Approach

### 1. INTRODUCTION

Over recent years, much obvious and immediate damage to marine ecosystems, or their components, has occurred, due directly to waste discharge and physical changes to habitats. At the same time, the process of degradation attributed to human activities has, in many cases, been very slow, and when damages are recognized it may be too late to intervene. The scientific community is often requested to identify or predict possible and/or probable long-term effects of low-level chemical contamination in order to give an early warning of such changes. This is a difficult task to undertake; on the one hand, ecosystems are highly complex and variable in time and space, and in most situations the absence of a "norm" or of baseline studies makes it impossible to establish the previous or pristine status of that particular ecosystem or community. On the other hand, there is the problem of identifying the source of contamination or the type of exposure. It is obvious that contaminants, usually multi-component, can interact or have different degrees of bioavailability and that exposure patterns can vary with time. This makes the examination of low-level contamination a challenging and complex task.

The evidence for and significance of long-term changes associated with the effects on marine biota of low-level contamination is frequently challenged by policy-makers, industry and scientists. A more structured conceptual and analytical framework is needed to document and test the evidence of change in marine biota as a result of contamination, to determine whether it is permanent or irreversible, and to identify a probable cause or causes. It is important to distinguish natural from anthropogenic effects, to select communities or ecosystems that reflect the slow build-up of contamination, to translate observed effects in individual organisms to their populations and communities, and to ensure that methods used for both sampling and analysis are suitable for the task (Carney, 1987).

It should be noted that subtle but important changes in ecosystems or communities are often equally difficult to measure. In addition, it should be accepted that change is not necessarily "damage"; the latter is specified in the GESAMP definition of pollution as a "deleterious" effect on, for example, marine resources and activities. Thus, damage may be judged subjectively or objectively according to each specific identified interest.

This report provides an analytical approach and tests its validity by reference to four 'case studies' representing quite different classes of contaminants. Its aim is to stimulate a wider audience to consider other cases that may support or contradict the underlying hypothesis that low-level contamination or long-term exposure to accumulated residues results in measurable adverse consequences for biological components of the marine ecosystem.

#### 2. PRINCIPLES AND CONCEPTS

It is necessary to define what is meant here by the terms "long-term" and "low-level". For "long-term" one might refer to arbitrary definitions such as geological scale, life-cycle of a single organism or the life-span of the longest lived species in an ecosystem (Connel and Sousa, 1983). It is proposed here to relate "long-term" to human life, in particular to a time scale of one generation. Some negative effects have already been identified on this time scale, eutrophication, for instance. Appropriate timescales could be shorter, i.e. TBT is slowly released from paints applied to ships on a more rapid time scale of months and ecological responses are evident within a few years. problem must, therefore, be approached with some flexibility. The definition of "low-level" needs to be considered equally on a case-by-case basis, paying attention to the chemical and toxicological characteristics of the substances of interest. However, the crucial point in the selection of cases to be used in this report is to document the contaminant "load". In most of the cases considered we observe "low" concentrations but heavy loads (i.e. high total quantities) of a single contaminant or of a mixture of substances (e.g. in estuaries of big rivers) which involve exposure of the ecosystem to low concentrations of single contaminants but a high degree of total contamination. In many cases, it may be impossible to trace and identify the relevant substance or substances that could cause certain effects. While the effects of mixed contaminants at acute effect levels are generally taken to be additive, lower concentrations may make a lesser contribution to an overall effect (EIFAC, 1987).

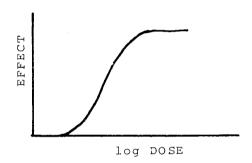
Nevertheless, it has been demonstrated that chemicals, especially organic compounds, belonging to similar 'chemical groups' (i.e. similar mode of action, same QSAR (Quantitative Structure-Activity Relationship) equation; Könemann, 1981) are additive at very low concentration levels. In predicting long-term effects of low-level exposure, attention must therefore be paid to the question of the "total load".

Moreover, at least two situations should be distinguished: (a) exposure for long periods to low levels of trace contaminants and (b) the slow accumulation of a contaminant due to its persistence, building up to a level that provokes effects.

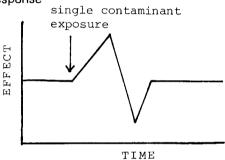
The theoretical basis for the approach taken in this report is derived from concepts of toxicology applied to environmental problems. The classic dose-response relationship relates mortality or some other measured response to an exposure (dose) or a concentration which prevails for a specified time.

The general patterns of dose-response are shown in Figure 1.

### 1. The classical dose-response relationship



### 2. Incorporating the time component/individual response

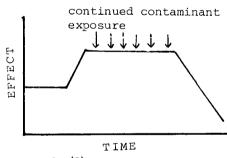


Response:

Metallothionein induction (metals)

MFO activity (PAH, PCB) Respiration (general)

### Response to repeated or continued exposure



Response:

As (2), or

Eggshell thinning (DDT)
Imposex in Nucella (TBT)

\_\_\_\_\_

Response to low-level contamination

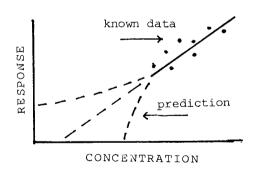


Figure 1. Patterns of dose response

Many data exist from laboratory or field exposures to show the dose-response relationship at higher concentrations or exposures. However, this relationship needs to be examined for low levels at the lowest part of the curve where responses may be less obvious or where predictions of deleterious effects are required in advance of clear evidence (Fig. 1.4).

Another factor that increases the complexity of the problem is the response that an organism, population or community will give when an additional stress factor is present in the presence of a low level of a contaminant.

Application of a single dose of a toxic substance at non-lethal concentrations will usually lead to a measurable response in an organism which, after a recovery period, returns to normal function (Fig. 1.2). The recovery response is often to ameliorate the effect of the toxic agent, as, for example, heavy metal exposure leads to production of metallothioneins which counteract the initial toxic response. For PAHs and PCBs, mixed function oxidase enzymes are activated to counteract the stress response, and for organophosphorus compounds, cholinesterase production is inhibited. Less specific, but common, responses such as changes in respiration rates or in growth are well documented for fish and bivalves. All these responses can be used as "biological response tools" for monitoring the effects of the applied agent. In some cases, where the responses are specific, they can be used to identify the type of agent involved and in developing an "early-warning" strategy.

If a target organism or ecosystem is exposed continuously to a toxic agent, the response will often be shown continuously and may, as a consequence, lead to severe reproductive failures, with effects on populations and communities. For example, reduced fecundity in bivalves is a consequence of reduced "scope for growth", and egg-shell thinning in sea birds exposed to DDT and PCB residues in diet, or induction of "imposex" in the whelk, Nucella, exposed to TBT, all result in changes in population levels. Community level responses in benthic assemblages exposed to organic enrichment have also been known for a long time (Pearson and Rosenberg, 1978).

From these considerations, we can argue that at concentrations of toxic substances commonly found, and measured, in contaminated environments, we can observe responses in individuals, populations and communities. The critical question is whether these, or other, effects occur at lower exposures than those known, i.e. where the response curve in Figure 1.4 is only predicted (dotted line).

Notwithstanding the complexity of this problem, some indication can be given on possible biological early-warning signals of deleterious effects. For example, a community often shows graded responses to non-specific contamination in a step-wise progression, such as:

- (i) rare or sensitive species are absent,
- (ii) quantitative changes occur in some species,
- (iii) reduced diversity occurs,
- (iv) dominance of opportunist species develops.

Some communities with high natural fluctuations, such as plankton, are likely to be less useful than those which are inherently more stable, such as benthos. Single species changes are more relevant if the species maintains its population by low fecundity and long generation time.

Finally, there are parallel early-warning signals to be derived from chemical information. For example, the presence of a potentially toxic chemical where it should not be, increased concentrations in water, sediments or tissues of naturally occurring substances or changes in ratios between elements, are all indicative of changes with possible deleterious biological effects. Accumulation of potentially toxic chemicals in biota has greater significance when a predator at the top of the food chain is involved. In addition, the linkage between predictions from fate models and monitoring chemical residues in environmental samples shows major promise as an early warning tool for contaminants (Clark et al., 1988).

### 3. CASE STUDIES

Examples can be selected to demonstrate that "long-term" exposures to "low levels" have caused effects that can be considered as damaging. To explore the validity of this thesis, some cases for which long time-series data exist can be studied. As a start to the analytical approach, it was considered helpful:

(i) to examine a few illustrative cases where there are sufficient qualitative and quantitative data;

- (ii) to define the best criteria to assess ecological damage;
- (iii) to select indicative responses for application to other examples and to guide monitoring strategies.

While observations that ecological change has occurred are quite numerous, it is not always clear that this can be attributed to an identified agent, particularly at low levels. On the other hand, there are some examples where a cause is indisputable or where there is clear evidence that the toxic agent is present in low and persistent concentrations. Some examples are given in outline in this section to illustrate the sequence of responses and to develop the principles on which other examples, possibly not so well documented or where a causal agent is only surmised, could be assessed. This approach, if useful, could be further developed to provide a prediction of long-term damage to ecosystems, its time scale and possible recovery, and to identify "early-warning" signals.

The examples selected here represent some diversity of contaminant type and of biological response so that the approach developed in later sections of this report should have some generality. They are well researched and documented, so that sufficient material is at hand for evaluation. They are as follows:

- (a) nutrients (nitrogen, phosphorus; primary productivity)
- (b) chlorinated hydrocarbons (persistent xenobiotics, e.g. DDT; reproduction)
- (c) tributyl tin (toxic chemical; growth, reproduction)
- (d) hydrocarbons (persistent mixtures; wide range of less specific biological responses).

#### (a) Nutrients

- 1. Primary productivity and standing stock biomass in the marine environment are regulated mainly by levels of nutrients and by physical factors such as temperature and light. Phosphorus (P) and nitrogen (N) play the most important role, one or other often being the key factor controlling primary production and biomass. In the last 20 years, eutrophication, defined as "enhanced nourishment leading to stimulation of aquatic plant growth by mineral nutrients, particularly the combined forms of P and N (GESAMP, in press)", has increased world-wide in the freshwater environment, and more recently several enclosed seas or parts of oceans began to show the same phenomenon. The Adriatic and Baltic Sea are two examples; in the former, primary production was regulated (limited) mainly by P, and in the latter by N.
- 2. Eutrophication of marine waters is a clear case of the long-term effects of low-level exposure where, despite the limitations described in Section 2, several points have been clearly established. For example, off the Dutch coast, between 1930-80, N has increased by a factor of 4 and P by a factor of 2 (Postma, 1978; United Kingdom, 1987), whereas off the German coast, over a period of 23 years, nitrate-N increased by 1.7 and phosphate-P by 1.5. In the German Bight phytoplankton biomass has increased dramatically over the same time period, with flagellates increasing sixfold whereas diatoms have been halved (calculated on basis of changes in C content).
- 3. In these examples there is an implied relationship between nutrient concentration and the amount of primary productivity and standing stock biomass, but it is nutrient flux and not concentration that is the critical factor. For example, in the Oslofjord, Norway, in summer, zero concentrations of N are recorded, yet phytoplankton production is high because the cells are able to utilize rapidly any ammonium excreted by zooplankton (Paasche and Kristiansen, 1982). Thus, in establishing dose-response relationships between phytoplankton production and biomass with nutrient inputs, fluxes are relevant rather than concentrations in unit time or area, i.e. quantities of nutrient available.
- 4. A secondary consequence of eutrophication includes changes in species composition (flagellates dominate rather than diatoms), the production/respiration ratio is inverted, leading to anoxia and mass mortality of many species (e.g. off the German coast in 1981 and in the Kattegat in 1985, 1986, 1987) and benthic communities show increased biomass and a change from dominance by suspension- to deposit-feeders (Pearson and Rosenberg, 1978).
- 5. In the Skagerrak in May 1988 a huge bloom of the algae <u>Chrysochromulina polylepis</u> occurred. Densities up to 72 million cells per litre were recorded above the pycnocline at 10-12 m depth.

Whilst there is no general agreement as to the causes of the bloom, the amount of N incorporated within the cells was up to 20 times the amount normally occurring in the whole water column, suggesting that excess N was a key factor.  $\underline{\text{C. polylepis}}$  released toxins affecting cell membranes and thereby osmoregulation processes leading to massive fish kills in both wild and cultured species, and to high mortalities in a variety of benthic species found down to 12 m depth along over 200 km of the west coast of Sweden and the south coast of Norway.

- 6. Thus there is clear evidence of significant changes in the inputs of nutrients and in increased primary production and changes in benthic biomass. The causes are identified as increased anthropogenic nutrient inputs. There is a sequence of effects, firstly an increase in production and biomass, secondly a change in species composition in plankton and benthos, and thirdly, in extreme cases, anoxia and high mortalities. The significance of the ecological changes is important in that, although initially eutrophication leads to increased biomass and is a potential source of fish food, the change to increased respiration and anoxia is an abrupt and catastrophic process. Determining the dose-response relationship and the level at which this change occurs is of primary importance and has not been established in the sea. Production of toxins during blooms is also important ecologically and may be a secondary effect of eutrophication with implications for human health.
- 7. Plankton blooms are transient phenomena in coastal waters receiving excess nutrient inputs. Thus eutrophication is reversible. Well documented case-histories of reestablishment of natural benthic communities following cessation of the input are known (Pearson and Rosenberg, 1978).

### (b) Chlorinated Hydrocarbons

- 1. Synthetic insecticides such as DDT, PCCs (toxaphene) and other chlorinated compounds (e.g. PCBs, chlorophenols) have been widely used in western countries for several decades. While their use in developed countries has declined, in developing countries it is still increasing. Further, even though their use in some areas has stabilized or declined, persistent residues of these materials are still being recycled, for example, from sediment reserves through the lipid pool within the plankton, so providing a persistent source of low-level contamination. Typical levels in Baltic marine sediments are now: DDT 9.9 mg/kg lipid and PCB 4.1 mg/kg (Reutergardh, 1988).
- 2. The hydrophobic and lipophilic nature of organochlorines means that in the marine environment they are more concentrated in the surface layer, along with nutrients and other organic materials. In this surface layer the high level of biological activity incorporates organochlorines into the food chain. The accumulated material is transferred progressively to higher trophic levels, leading to biomagnification. Thus plankton contains 0.25 mg DDT/kg lipid, gulls (Larus ridibundus) 75 mg/kg and the sea eagle (Haliacetus albicilla) 140 mg/kg. For PCBs the concentrations are 0.55, 150 and 340 mg/kg lipid respectively (Reutergardh, 1988).
- 3. Although the materials were well characterized chemically and methods of analysis are now reasonably well developed, damaging effects in the environment were not established for some decades. Monitoring of organochlorine residues in food items (e.g. fish flesh) is directed to human health protection; monitoring of other environmental components is done to establish distribution and temporal trends.
- 4. In the fifties a drastic decline in populations of birds of prey was seen and associated with high organochlorine levels in their tissues. At that time, a relationship between DDE concentrations in bird tissues and in egg-shell thickness suggested that reproductive failure could be the cause of population decline. Only after many years of PCB use were threatening levels of PCB in fish and predatory birds reached. Different species were shown to have different sensitivity.
- 5. Increase in production and use of DDT and PCBs was followed within about 5 years by increase in their concentrations to about 600 mg/kg (lipid) in Baltic guillemot (<u>Uria aalge</u>) eggs, but have since declined over a decade with reduced use to previous levels. Shell thickness has been shown to be inversely related to DDE concentrations in the eggs. Only after a decline to less than 200 mg/kg (lipid) did shell thickness revert to normal values. Recent parallel fluctuations in both are not yet well explained (Reutergardh, 1988).
- 6. The dramatic decline in grey seals (<u>Halichoerus grypus</u>) in the Baltic Sea has also been related to accumulation of high concentrations of organochlorine residues which affect reproduction (uterine occlusions). However, there appears to be no firm conclusive evidence linking residue

concentrations in marine mammals with effects on reproduction (Addison, 1989). The recent high mortality of North Sea common seals (Phoca vitellina) has been linked to virus infection (Harwood and Rijnders, 1988). The possibility of a weakened immuno-response mechanism associated with PCB exposures exists. In other studies levels of PCB residues in the flounder (Platichthys flesus) have been related to increased enzyme activity (Stegeman et al., 1988). While this indicates increased exposure, it is not known if the health of the population is affected or the incidence of fish disease increased.

7. Thus there is evidence of a change in populations of some top marine predators (e.g. seals, peregrine falcons or sea eagles) as a result of slow build up of persistent organochlorine residues in the marine environment. Changes in organochlorine use are reflected in levels of residues but the effects on target species are not so clear; a threshold limit for known effects (e.g. egg-shell thinning) is recognized but observations of effects at much lower levels are of unknown ecological significance. The recycling of persistent residues within polluted systems means that biological recovery will be slow or unlikely if these low levels are important and effects may not match changes in use if there is a time-lag of response.

### (c) Tributyl Tin

- 1. Organotin compounds have been developed in recent decades and have been used increasingly as antifouling and biocidal agents in the marine environment. They are used in the formulation of marine paints and to treat materials used for nets and cages. Although some organotin compounds are also used in the terrestrial environment, their low mobility in air and sediments and rather rapid degradation means that this use is unlikely to lead to a significant marine problem. Much of the information in this profile has been taken from a recent UNEP/FAO/WHO/IAEA review (1988). Another independent, review is that of Champ and Pugh (1987).
- 2. Organotins used in the marine environment are forms of butyl tin, especially tributyl tin chloride and oxide. These compounds dissociate in sea water, forming chloride and hydroxides. It is probable that residues in the marine environment can be methylated by biological or abiotic routes. These compounds are all lipophilic chemicals of low volatility, and they tend to accumulate in sediments and the biota.
- 3. TBT compounds are generally not very persistent with degradation times about 7 to 15 days for compounds taken up by marine biota and about 160 days for marine sediments. This means that if input is stopped, contamination can be lost in a relatively short time (i.e. within 1 year) but continued use and progressive leaching from antifouling treatments provide a slow and persistent release at low concentrations (picomols/1).
- 4. Acute toxicity has been demonstrated for a wide variety of marine organisms at levels of a few  $\mu g/l$ . Such acute toxicity studies are of value in establishing the relative toxicity of different compounds and the comparative sensitivity of organisms and life stages. Toxicity increases with the length of the alkyl chain, and larvae and juveniles are generally more sensitive than adults.
- In longer-term laboratory tests abnormal shell calcification and reduced body weight and growth in oysters, and limb deformities in crabs and starfish, were observed at concentrations from 1 to 0.01  $\mu$ g/l. A decrease in density of several species exposed in a microcosm experiment was observed at 0.5 to 1.8  $\mu$ g/l. Dog whelks (Nucella lapillus) developed "imposex" (the anomalous development of male characteristics) at about 1  $\mu$ g/l exposure concentration. Imposex is the most sensitive effect recorded for TBT, and for this reason it has been used as a field bioassay to measure concentration-related effects in estuarine and coastal environments.
- 6. Field data from the Arcachon Bay, France, indicated that TBT released from antifouling paints caused serious abnormalities in the shell calcification of the Pacific oyster <u>Crassostrea gigas</u>. In U.K. waters, imposex was observed in female mud snails <u>Nassaria obsoletus</u> and in female dog whelks, Nucella lapillus.
- 7. Concentrations of TBT found in such "hot spots"  $(\geq 1~\mu g/l)$  are likely to have an acute toxic effect on sensitive marine organisms. These effects may reduce the diversity at affected sites by loss of sensitive species, especially if these are not able to recolonize areas where pollution has occurred. Growth reductions are observed for a wide range of organisms.
- 8. There is clear evidence that the use of TBT in antifouling agents is responsible for adverse effects leading to loss of species in local areas. A sequence of effects from growth anomalies

through to reproductive failure is observed; a dose-response relationship is evident. Continued use of TBT products is likely to have extensive effects both ecologically and commercially. Observation of effects in the field have led to identification of the cause and to rapid control of TBT use.

### (d) Hydrocarbons

- 1. Mesocosm and field studies, and studies at accidentally-oiled sites have shown that a wide range of sublethal biological effects occur after marine organisms and their communities are exposed to petroleum hydrocarbons at low concentrations. The literature has been recently summarized by Clark et al. (1982), U.S. National Research Council (1985, 1989) and Boesch and Rabalais (1987).
- 2. Mesocosm studies have been conducted in several laboratories. In experiments at lower (sublethal) concentrations, scope for growth in Mytilus edulis showed a decline at concentrations of water-accommodated fractions of diesel oil as low as 20 μg/l (Widdows et al., 1982). At the community level, reductions in diversity and changes in size structure of phyto-and zooplankton occurred in oiled tanks at concentrations down to 75 μg/l (Grice and Reeve, 1982; Gamble et al., 1977). Meiobenthic copepods but not nematodes nor macrofauna showed differences at dose levels down to 3.4 + 1.8 μg/l diesel oil over a period of 20 weeks (Warwick et al., 1989).
- From recent field surveys at the sub-lethal level, enzyme activity in flounder (Platichthys flesus) was positively correlated with a gradient of PAH in Langesundfjord, Norway (Addison and Edwards, 1988; Stegeman et al., 1988). Scope for growth in M. edulis was also positively correlated with the gradient of PAH there (Widdows and Johnson, 1988). In these studies, however, other contaminants may have been present.
- 4. Continuous low-level HC contamination occurs near operating oil platforms. Data on the North Sea show that benthic community diversity is significantly reduced at total HC concentrations in the sediment of 100 mg/l, and reduction in diversity is indicated at ≥ 10 mg/l (Davies et al., 1984; Reiersen et al., 1989).
- 5. Long-term studies at several oil spill sites (e.g. FLORIDA, Buzzards Bay, 1969; AMOCO CADIZ, 1978) where hydrocarbons have remained in the sediments subtidally or in the intertidal zones, have shown, amongst others, effects from residue accumulation on behaviour/recruitment in crabs of Buzzards Bay and the Brittany coastline and successional changes in benthic amphipod populations (Buzzards Bay). Vandermeulen (1982) describes long-term (up to a decade) biological and ecological effects from marine oiling. More recently, sublethal effects were observed at low concentrations in sub-tidal sediments after 18 months at an oil spill site in Panama (Jackson et al., 1989).
- 6. There are therefore many recorded changes both sub-lethal and lethal in response to HC contamination, some at low concentrations. There are clear links between contamination levels and biochemical responses, and effects on physiological processes, populations and communities. However, the extent of effects on benthic communities around North Sea oil platforms covers only 2-3% of the total area of the North Sea (Reiersen et al., 1989), and damage is reversible if contamination stops. As yet, the dose-response relationships cited are crude and must be further refined.

On the basis of these profiled case studies, recorded effects can be summarized as in Table 1. Effects can be characterized and dose-response relationships established which range through all levels of biological organization. Other possible indicators include size shifts in communities, genetic and CMT (carcinogenic, mutagenic, teratogenic) effects and metallothionein, but are not listed.

An examination of the four examples in Section 2 shows that in each case sufficient information is available to select an appropriate response target, to identify and measure the putative cause, to judge sensitivity to other interacting factors, to develop correlations, trends and causation. Methods of sampling/measurement and analysis are sufficiently reliable. From such documented profiles the overall ecological significance can be assessed with some confidence.

Of course, the examples selected here are lacking in detail. Further information is available to justify the conclusion that at least in these examples long-term and widespread ecological changes can be reasonably attributed to low persistent concentrations of contaminants or their slow build-up in the marine environment.

Table 1

Examples of biological response to four contaminants at low concentration

### Indicators of Biological Response

Case	Mixed function oxidase (MFO)	Bio-concentration (BCF)	Scope for growth	Production/ respiration	Loss of species	Change in diversity	Reproduction failure	Secondary toxicity
Nutrients	*		?	•		•		•
Organochlorines	•	•	•		•	•	•	
TBT		•	•		•	•	•	
Hydrocarbons	•	•	•		•	•	•	

### 4. DEVELOPMENT OF AN ANALYTICAL FRAMEWORK

One or more analytical frameworks can be developed from the concepts set out in Section 2 for an objective and critical evaluation of the case studies profiled in Section 3 to evaluate, as clearly as possible, the evidence of long-term biological and ecological responses to low-level contaminant exposure.

Two possible frameworks follow. The framework approach is intended at this stage as a working procedure for assessment of contaminants of different character using the data presented in the "case-study" profiles to show how the approach might be employed but they are neither comprehensive nor definitive. If this approach proves useful, we believe that it could be developed further and applied to other classes of contaminants.

These frameworks should not be regarded as a manual for contamination assessment but simply as guides for the case-by-case consideration of the evidence needed to clarify the problem. The use of MATRIX ONE is demonstrated by two of the case studies in Section 3. MATRIX TWO is exemplified by information from results of a joint investigation (sponsored by IOC/GEEP) of biological effects of contaminants on a selected number of marine species and communities (Bayne et al., 1988).

Together, the two matrices can guide the critical selection and analysis of these and other case studies and ensure that both "low-level contaminant exposure" and "ecological effects measurements" are satisfied in each case.

Each framework, with its elements, should allow us to

- (a) judge the current technical and theoretical ability to detect changes induced by low-level exposures, with confidence;
- (b) evaluate the characteristics, strengths and weaknesses of the unequivocal evidence showing such contaminant-induced change;
- (c) evaluate the case histories/data sets of changes in marine ecosystems that might be, in part, due to low-level contaminant exposure, but where such changes are masked by natural factors and complexity, or by data limitations, and
- (d) identify a putative agent and establish a dose-effect relation by observations and testing.

MATRIX ONE (Table 2) is organized as a contaminant exposure assessment, considering the factors influencing contaminant presence and distribution on a broad marine ecosystem basis. This simple matrix provides a means of categorizing and summarizing exposure to a contaminant according to its nature and distribution. In the case of N and P nutrients, for example, there is little evidence of an excess over natural concentrations in oceans, but many examples of increased levels or inputs in inshore waters. Both concentrations of N and P, and "loads" from discharges are usually known and at the primary production level exposures could be expected to lead to increased biomass. However, this will be less evident at the secondary trophic levels. Similarly, DDT and other organochlorines can be identified and reasonably quantified in sediments, and through bioaccumulation reach high levels of exposure in top food-chain predators. These examples are used in completing the matrix, showing where the contaminants are likely to be found in both non-biological and biological compartments.

MATRIX TWO is organized as an effects assessment, considering factors important to ecological functions (Table 3). It simplifies assessment of biological responses by categorizing the nature of the response and the level of organization at which it operates. Further, it indicates whether these responses are amenable to routine measurement of contamination and documentation of ecosystem response. At the different levels of biological organization it identifies robust and reliable techniques for both generalized and specific responses.

Despite limitations, including the small number of cases described, the matrices aim to provide a logical scheme for an objective procedure for any case for examination.

Table 2

MATRIX ONE. A check list of contaminants:
Characteristics, distribution and fate

	Non-biological compartment			Biological compartment			
Contaminant Characteristics	Oceanic waters	Inshore waters	Sediments	Plankton	Benthos	Fish	Mammals and birds
Identification		•		•	0	0	
Quantification (concentration and "load")		•		•	0		
Increased natural substances							
Xenobiotic substances							
Single contaminant							
Mixed contaminants							
Gradients and clusters (spatial)		•	0				
Persistence	ļ	0		<u>.</u>			
Accumulation							
Secondary toxins		•		0		•	
Widespread distribution							
Longtime scale		0					

The matrix is applied here to two contaminants from Section 3, nutrients (N & P) $\odot$ , and DDT/PCB  $\Box$ , with reference to the evidence available in each case

Level of occurrence: • = high • = medium • = low