

IMMEDIATE IMPACT OF *AMOCO CADIZ* ENVIRONMENTAL OILING:
OIL BEHAVIOR AND BURIAL, AND BIOLOGICAL ASPECTS

by

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RESUME

Les différents états d'altération du pétrole, depuis les huiles jusqu'aux mousses, ont des effets significatifs sur les interactions pétrole-eau et pétrole-sédiment lors de leur échouage. L'observation des plages polluées suggère que deux mécanismes interviennent lors de la contamination : la contamination et la pénétration de la couche superficielle par les huiles, et l'enterrement de couches distinctes par le purgeage des nappes de mousses. De grandes quantités de pétrole échoué peuvent se retrouver logées dans les sédiments de la plage selon le temps de contamination et la dynamique de la plage. Elles restent disponibles pour un relarguage ultérieur.

La toxicité létale aigue est intermédiaire à celle du pétrole brut Bunker C et du Koweït. Mais des études d'altération simulée suggèrent, cependant, que la toxicité aigue augmente directement avec l'altération.

Les mortalités dans la zone intertidale ont été très variables, quelques espèces ont subi une mortalité presque totale tandis que d'autres, notamment les annélides et les némertides, ont été apparemment très résistantes à la contamination. Les oiseaux et les poissons pollués avaient des concentrations d'hydrocarbures et des activités d'aryle hydrocarbure hydroxylase beaucoup plus élevées que ceux qui n'étaient pas pollués. Les conséquences de l'altération du pétrole sur la toxicité des nappes échouées et des nappes enterrées sont discutées ainsi que les mécanismes de défense possibles écologiques ou physiologiques.

ABSTRACT

Weathering states of oil ranging from sheen oil to mousse have significant effects during oil stranding on oil-water and oil-sediment interaction. Observations of oiled sandy beaches suggest two mechanisms of beach contamination - general penetration and contamination of beach substrate by sheen oil,

and burial of discrete layers resulting from entrapment of mousse slicks. Depending on timing of oiling with respect to beach dynamics, large amounts of stranded oil can be accommodated in beach sediments. These are then available for later release.

Acute lethal toxicity of stranded *Amoco Cadiz* oil is intermediate to Bunker C and of Kuwait crude. Simulated weathering studies suggest, however, that acute toxicity increases directly with weathering.

Mortalities in the intertidal zone were highly variable, some species experiencing near total mortality while others, notably burrowing annelids and nemerteans, appeared highly resistant to oiling. Hydrocarbon concentrations and aryl hydrocarbon hydroxylase activity of oiled birds and oiled fish were found to be markedly higher than those in non-oiled species. The implications of weathering on toxicity of stranded and buried oil, and possible ecological and physiological defense mechanisms are discussed.

INTRODUCTION

The *Amoco Cadiz* incident (March 16, 1978) presented a unique opportunity to examine under prevailing spill conditions certain immediate short-term aspects of oil impact - the behavior and toxicity of freshly-spilled crude oil in an inshore marine environment. Of particular interest were the behavior of freshly-spilled oil during mousse formation and oil-burial in porous sandy sediments and the potential re-release of such entrapped petroleum hydrocarbons. Although this aspect of oiling has been examined to some degree (e.g. Thomas, 1973, 1978; Owens and Drapeau, 1973; Owens and Rashid, 1976; Owens, 1977, 1978; *viz.* also Environmental Protection Service, 1977), most studies to date have dealt with the post-spill situation, while in others conclusions have been drawn from simulated laboratory investigations. Thus, while burial of oil in beaches following a spill has been documented (Anonymous, 1970; Vandermeulen and Gordon, 1976; Owens and Rashid, 1976), the significance of buried oil in terms of beach dynamics has not been fully appreciated.

Similarly, although many toxicity studies have been carried out, the toxicity of a freshly-spilled oil during initial natural weathering and the biological tolerances are not well understood.

In this manuscript we describe observations and preliminary conclusions resulting from studies carried out within four weeks of the initial grounding of the *Amoco Cadiz*, and while fresh slicks were still coming ashore along the north shore of Brittany.

MATERIALS AND METHODS

Samples (sediments, oil, biota) were collected in chemically clean glass jars (rinsed with double-distilled solvents) and stored at -10°C . For analysis at the Bedford Institute of Oceanography in Dartmouth, Canada, samples were transported in dry ice, and subsequently stored at -20°C until analyzed.

Oiled birds were obtained from a bird-cleaning clinic through the assistance of Monsieur J-Y Monat of the Université de Bretagne Occidentale at Brest, France. Severely oiled birds assessed to be beyond recovery were sacrificed and the freshly collected tissues were frozen and stored as outlined above.

Oil samples from the *Amoco Cadiz* were not available. Therefore, samples of freshly stranded oil were taken for reference.

Sediment Hydrocarbons

Following thawing, known amounts of sediment samples were extracted by the method of Farrington and Tripp (1975), with minor modifications as described in detail by Keizer *et al.* (1978).

Hydrocarbon concentrations were determined spectrofluorometrically (Vandermeulen *et al.*, 1977). Synchronous scans (Hargrave and Phillips, 1975) were also done periodically for further hydrocarbon identification.

Tissue Hydrocarbons

Extractions were carried out by the methods of Gordon *et al.* (1978), with only minor modifications. Hydrocarbon concentrations were determined spectrofluorometrically, and were expressed as micrograms of *Amoco Cadiz* oil equivalents per milligram protein nitrogen (Lowry *et al.*, 1951). Characterization of tissue aliphatic hydrocarbons was done on pentane extracts using an HP-7620 gas-liquid chromatograph (FID, 3% Dexil 300 on Chromosorb W HP, 6°C min⁻¹).

Oil Toxicity

Acute toxicity, expressed as 2-hour LC₅₀s, was estimated for the flagellate unicellular alga *Monochrysis lutheri* (Vandermeulen and Hemsworth, 1977) which were exposed to aqueous extracts of oil. Extracts were prepared by shaking 5 mL oil in 250 mL filtered sea water (0.45 µm, Millipore). The resulting mixture was allowed to separate in a glass separatory funnel for one hour, after which aliquots from the lower aqueous phase were added to a number of test tubes containing samples of the algal culture. Test concentrations of oil in water ranged from 0.01 ppm to 100 ppm.

Aryl Hydrocarbon Hydroxylase (AHH)

Enzyme activity was assayed by monitoring hydroxylation of benzopyrene to 3-OH-benzopyrene (Nebert and Gelboin, 1968). Results were expressed as picomoles of 3-OH-benzopyrene min⁻¹ mg protein nitrogen⁻¹, using purified 3-OH-benzopyrene as standard.

RESULTS

Oil Behavior

(a) Oil States

Oil impinging on the shoreline was present in at least three readily identifiable stages of weathering - sheen, early mousse, late mousse. Sheen oil, under the influence of various weathering conditions, will readily form into mousse by incorporation of water into oil. Late mousse differs from early mousse by having an increased water content together with increasing sand incorporation.

The meteorological and oceanographic conditions which prevailed at the time of the grounding, i.e. storm wave conditions with large amounts of suspended sediment during the beach constructional phase, greatly favored emulsification, with rapid changing from sheen oil to mousse formation. On the other hand, mousse resulting from the *Amoco Cadiz* oil appeared to be quite unstable, with ready reversal from mousse to sheen. Thus in several stranded oil pools, as in the Ile Grande salt marsh, a layer of relatively fluid black oil was found overlying the characteristic chocolate-brown mousse, suggesting 'breaking' of the water-in-oil emulsion.

Oil sheens covered large areas of water and were largely wind-driven. Sheens concentrated along the shoreline were deposited over the complete tidal

range during the ebb. Sheens were also occasionally visible as discontinuous patches on intertidal flats, particularly on flood-dominated areas, where sheen oil became available for penetration into porous sediments (e.g. at Grève St. Michel near Lilia).

Mousse, due to its increased viscosity with increasing water-in-oil concentration, penetrates poorly into porous sediments. Early mousse clings readily, and typically coated marsh grass (e.g. Aber Benoit, Ile Grande salt marsh) and beach sediments. Because of its clinging property it does not lift off readily during the following flood tide, and is therefore apt to become buried.

Late mousse, i.e. fully emulsified water-laden mousse, continually accumulated sand until the density of the resulting mixture exceeded that of water. Such mousse/sand mixtures tended to clump, and were moved and deposited preferentially into the more quiescent subtidal zones, such as near Portsall.

(b) Sediment Penetration

Examination of a number of sandy beaches showed oil incorporation into sediments of two types - more or less homogeneous contamination of sandy sediments versus burial of discrete oil layers within sediments.

The first, relatively homogeneous contamination of the sand column was observed on several sandy tidal flats near Lilia. Oil in sand concentrations averaged around 20.2 ppm in the upper oxidized layer and *circa* 5.8 ppm in the lower reduced sediments. Concentrations of petroleum hydrocarbons in the surface layer sediment were generally higher, averaging 32.4 ppm (Table 1).

Table 1. Petroleum hydrocarbon concentrations in beach sandy sediments oiled by *Amoco Cadiz* cargo oil. Means and standard deviations are presented.

Sample Depth	Sediment Hydrocarbons (ppm)*	Worm Cast Hydrocarbons (ppm)*
Surface	32.4 ± 20.7	-
Subsurface (5 cm) oxidized	20.2 ± 4.7	23.1 ± 4.8
Subsurface (15 cm) reduced	5.8 ± 5.9	0.7 (N=1)

* Hydrocarbon concentrations in un-oiled reference sediments averaged between 2.5 and 3.0 ppm.

Beach oiling by burial of discrete oil layers was observed in a number of beaches, most notably Treompan beach near Portsall (Fig. 1), Grève St. Michel near Lanion (Fig. 2), the mouth of the Aber Benoit near Portsall (Fig. 3), and the beach of the Centre Héliomarin near Roscoff (Fig. 4). Up to four and five distinct oil layers were observed at various depths below the sand surface, separated by apparently clean but contaminated sand layers. Oil concentrations

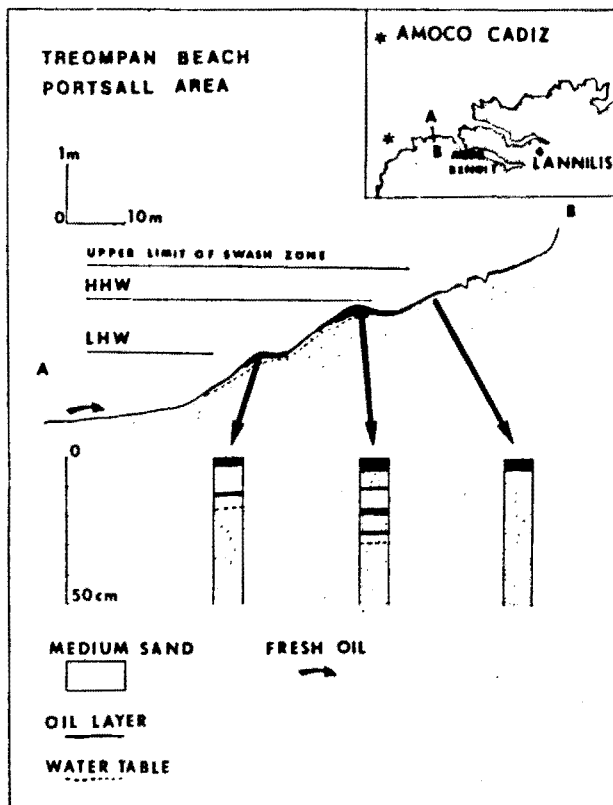


Figure 1. Beach profile of Treompan beach near Portsall, France, taken April 11, 1978.

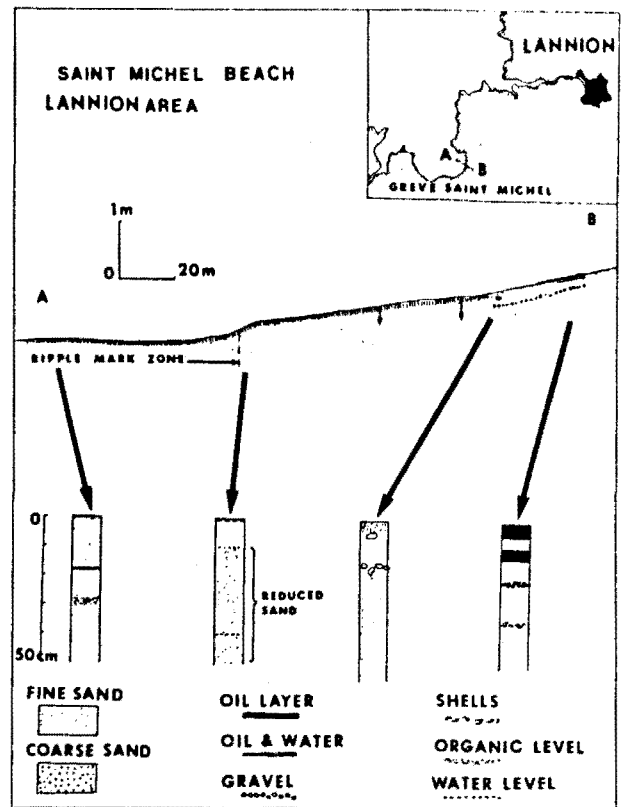


Figure 2. Profile of St. Michel beach near Lanion, France, taken April 11, 1978.

in the oily layers ranged from 72 to nearly 100 ppt in samples from the beach near the Centre Héliomarin. In contrast, the 'clean' sand layers, although oiled, were remarkably less oiled with hydrocarbon concentrations between 35 and 50 ppm (Fig. 4). Surface hydrocarbon concentrations in the upper one centimetre were slightly higher, averaging 185 ppm.

Oil Toxicity

(a) Two-hour LC₅₀

Two-hour toxicity tests using freshly collected *Amoco Cadiz* stranded oil indicated its acute lethal toxicity to *Monochrysis* sp. to be between that of Bunker C and Kuwait crude oil (Fig. 5). The two-hour LC₅₀ for the *Amoco Cadiz* sample was 4.4 ppm versus 3.3 and 8.6 ppm for Bunker C and Kuwait crude, respectively. Under similar test conditions the two-hour LC₅₀ for No. 2 fuel oil was 0.085 ppm, two orders of magnitude more toxic than the others.

The mortality curve for the *Amoco Cadiz* sample was similar to that for Kuwait crude with no detectable mortality over a two-hour assay at concentrations less than 1.0 to 2.0 ppm. Above 2.0 ppm mortality increased rapidly, with total mortality at above 10 ppm. The slope of the curve for the linear portion between 2 and 10 ppm was similar to that for Kuwait crude. That of Bunker C differed from these in being steeper.

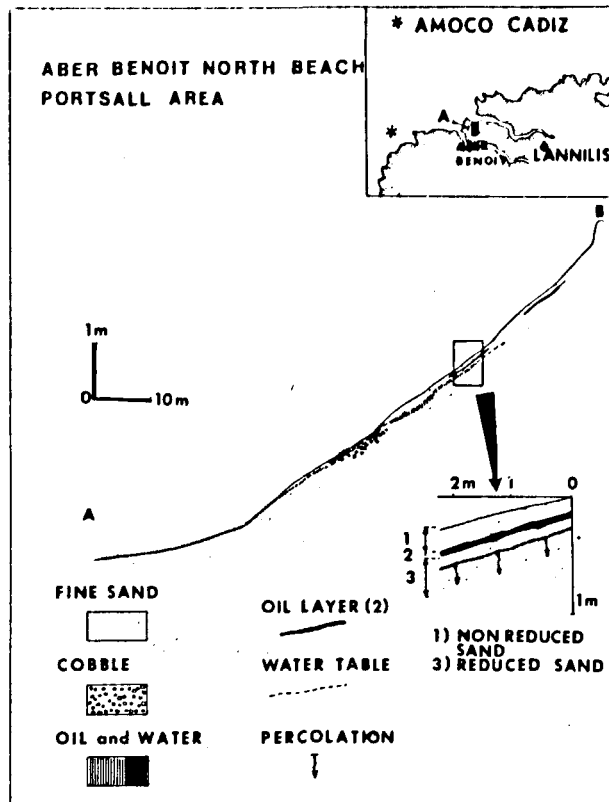


Figure 3. Profile of the beach at the mouth of the Aber Benoit, North Brittany, taken April 11, 1978.

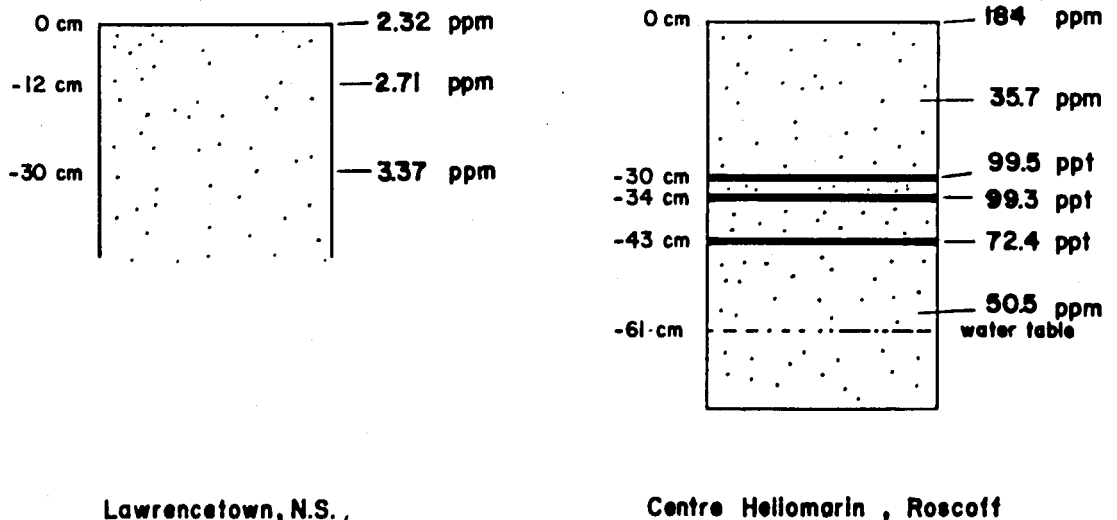


Figure 4. Section of trench in the beach of the Centre Héliomarin, Roscoff, showing oil in sand concentrations in parts per million (ppm) and parts per thousand (ppt). The adjoining section shows hydrocarbon concentrations found in un-oiled sediments from Nova Scotia, Canada. Concentrations were determined spectrofluorometrically.

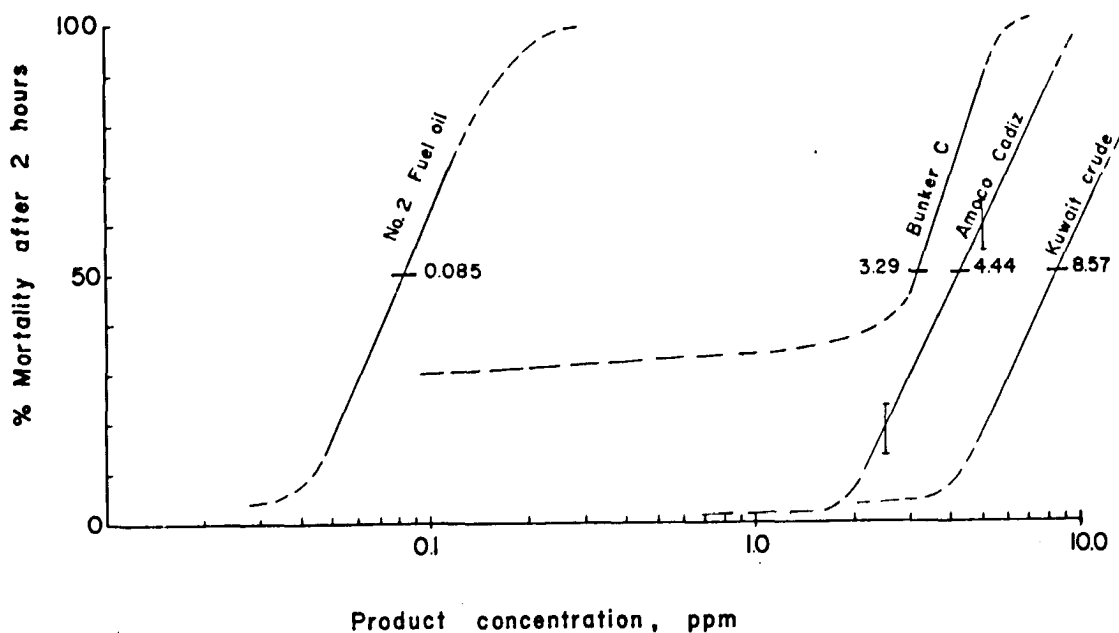


Figure 5. Toxicity curve for a number of crude oils and oil products using *Monochrysis lutheri* as test organism. LC₅₀ values are indicated by the small dash-mark for each curve.

(b) Weathering Effect

Preliminary results with laboratory-weathered oil (bypassing a stream of air over a sample for one week) suggest increased toxicity over time (Fig. 6). Also observed was a concomitant steepening of the linear portion of the mortality curve, similar to that seen with Bunker C.

(c) Biological Impact

All obviously oiled intertidal zones examined (sandy beaches, mud flats, rocky shores, salt marsh) showed heavy mortality of the intertidal macro-fauna communities although not all species were affected equally. High mortality was observed of the shore crab, *Carcinus maenas*, and the absence of several macroscopic organisms as amphipods and isopods in all oiled locations was noted. Heavy mortality of intertidal seaweeds, e.g. *Fucus* sp., was found in heavily oiled intertidal areas of Ti Saoson, an island offshore from Roscoff. In one location near the wreck of the *Amoco Cadiz* off Portsall lichens on rocks in the upper intertidal had been killed or severely damaged by sprayed oil.

Of the sandy beach in-fauna large numbers of normally sub-tidal cockles, *Cardium edule*, and Venus shells, *Venus* sp., were found in the intertidal on the surface of the sediments, either moribund or dead. In contrast, various burrowing species of polychaete annelids, including *Arenicola*, and nemerteans were found alive and producing normal worm casts. Large numbers of the heart urchin, *Echinocardium cordatum*, and of the razor clam, *Enis* sp., both dead and moribund, were found on the eastern end of the beach at St. Michel-en-Grève.

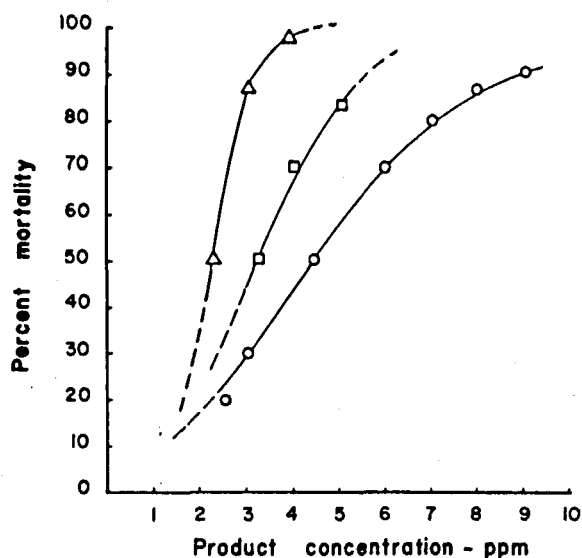


Figure 6. Effect of simulated weathering on toxicity of *Amoco Cadiz* oil. Open circle - freshly collected oil; open square - Bunker C fuel oil; open triangle - 'weathered' *Amoco Cadiz* oil.

On rocky shorelines sponges and sea anemones appeared largely unaffected by the oil, as were the common limpets *Patella* sp., despite quite heavy deposits of oil and mousse near the high tide line and obvious oiling of rocky tide pools. However, many species of marine gastropods, including the Littorinidae, were eliminated by all but very light exposures to oil.

Intertidally, all of the seaweeds were coated with oil, although the inner fronds seemed to escape heavy oiling.

Of the vertebrates examined schools of small mullet were seen swimming in the intertidal zone in a number of locations, including near the heavily oiled shoreline of the town of Lilia, and in marsh channels of the severely oiled salt marsh near Ile Grande. Dead fish were not found on any of the shorelines of north Brittany.

Aryl hydrocarbon hydroxylase (AHH) activity in mullet from the Ile Grande salt marsh was markedly higher than that normally associated with fish from non-oiled reference sites taken from Nova Scotia, Canada, waters (Fig. 7).

Analysis of oiled birds from north Brittany showed presence of petroleum hydrocarbons in all tissues from four bird species - liver, kidney, muscle, and brain. With the exception of loon liver all concentrations were greater than found in similar Nova Scotia bird species from non-oiled waters. In addition to the aromatic compounds detected by fluorescence, all tissues analyzed for aliphatics retained several aliphatic hydrocarbons (Fig. 8).

AHH levels in the oiled birds were generally higher than those found in non-oiled bird species (Fig. 7). Greatest AHH activity was found in the oiled

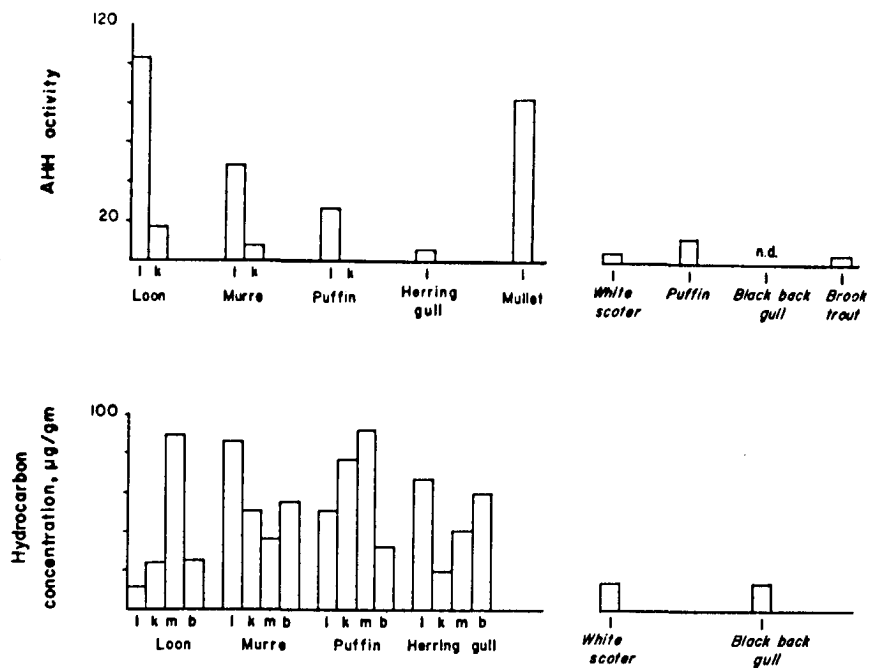


Figure 7. Aryl hydrocarbon hydroxylase activity and hydrocarbon concentrations in tissues from oiled (left) and non-oiled (italics right) birds and fish. l = liver, k = kidney, m = pectoral muscle, b = brain. Oiled birds and fish were collected in north Brittany four weeks after the *Amoco Cadiz* grounding. Non-oiled organisms were taken in Nova Scotia, Canada.

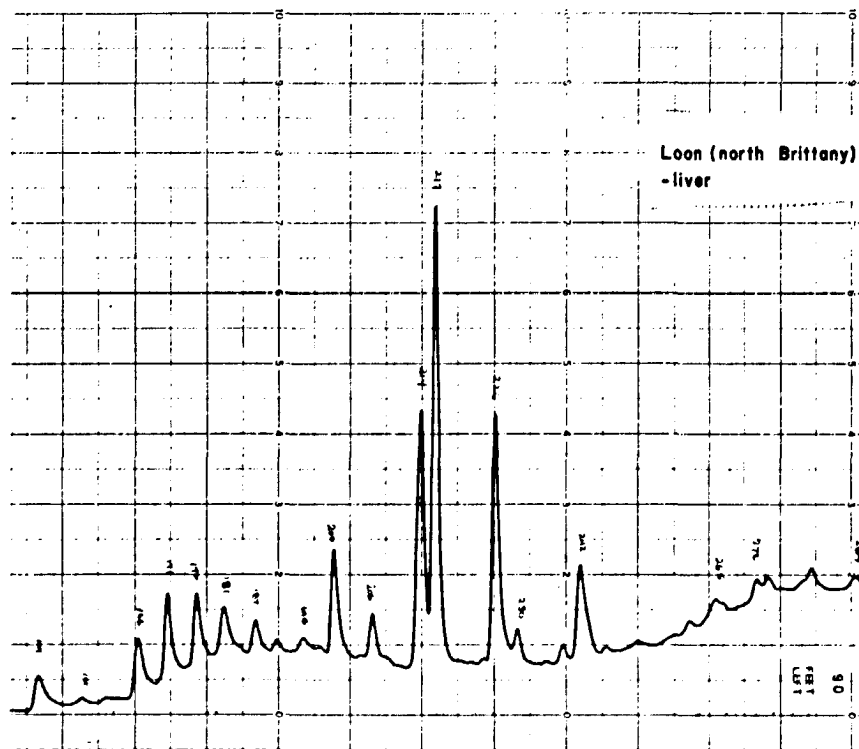


Figure 8. Gas liquid chromatograph spectrum of extract from oiled loon liver. Peaks at 21.4 and 21.7 retention times probably are of biogenic origin.

loon and murre, with lesser levels in the oiled puffin. That of the oiled herring gull was between that of control, i.e. non-oiled, puffin and control black-back gull.

DISCUSSION

Much of the immediate behavior of stranded oil results from its various weathering states, each state significant for subsequent oil-water and oil-sediment interaction.

Oil as sheen is readily available for dissolution into the water column, and because of its high mobility can readily penetrate the more porous sandy beach sediments. Because of its ready solubility sheen oil probably played a prominent role in the impact of the *Amoco Cadiz* spill on the subtidal and intertidal beach communities such as that off St. Michel-en-Grève, accounting for large mortalities of razor clams and heart urchins. Sheen oil was also probably largely responsible for the mortalities of *Venus* sp. and *Cardium* sp. near Lilia.

Sheen oil was also probably the main factor in the observed general contamination in the ppm range of sandy beach sediments, due to stranding of thin sheens on the tidal flats with the receding tide. The relatively high mobility of sheen oil together with tidal pumping of oiled interstitial water ensured thorough penetration throughout the beach structure. In this respect it is interesting to note that the reduced lower layers of the beach normally not accessible to oxygenation also appear to be less accessible to oiling via this pumping mechanism.

A potentially far-reaching consequence of the high mobility and solubility of sheen oil is that, by percolation through sandy sediments, hydrocarbons may reach the underlying water table, and thus be distributed considerable distances from the initial point of contact.

Stranded mousse, on the other hand, behaves entirely differently on sediments, tending to cling and to accommodate sand particles. As a result it contaminates by burial rather than by percolation. This was particularly evident in the case of the *Amoco Cadiz* where it is important to note that widespread pollution of the north Brittany coastline occurred immediately prior to the spring constructional wave activity, when sand which was carried offshore by winter storms was being returned to the beaches. This resulted in rapid burial of stranded oil, with interlayering of stranded mousse with new deposits of beach sand, resulting at some sites in up to five sequential buried slicks. (In fact, the situation in the beach of the Centre Héliomarin probably represents the results of the two processes superimposed on one another with mousse burial accounting for the observed oil layering, and the lesser, 30 ppm, contamination of the cleaner sand layers due to sheen oil penetration.) Throughout the summer constructional beach phase such buried oil layers will lie protected by the overlying sand layers, and oil buried in this manner is therefore relatively immobile - that is, until the onset of winter storm activity and the accompanying beach erosion. It is highly likely that at that time this buried oil will again be released into the marine environment.

The environmental corollary of this is that, whereas sheen oil acts in general oiling of sandy beaches in the ppm range, the constructional phase of beach geomorphology more specifically encourages the entrapment of mousse slicks in the ppt range. The amount of *Amoco Cadiz* oil entrapped through these mechanisms is not known. However, estimates from trenching studies suggest that this can be significant (*viz.* Fig. 4). Oil in buried mousse layers can reach concentrations of nearly 10% by weight of sand. Preliminary estimates suggest that as much as 7.5 kg of oil may be accommodated in this manner under one square metre of beach in the example cited. Thus, if even only 10% of the Centre Héliomarin beach were as described in Figure 4, a one-kilometre stretch of sandy beach *circa* 100 metres wide can contain up to 75 metric tons of stranded oil. By a very conservative estimate, assuming there are 12 similar beaches in north Brittany, some 900 metric tons or 4800 barrels of *Amoco Cadiz* cargo lie within the sandy sediments, ready for potential release at next winter's beach sand removal.

Little can be said on the initial acute toxicity of the original *Amoco Cadiz* crude oil, a mixture of light Arabian and light Iranian crudes, since no samples of the cargo were taken or were available. Any tests therefore were done on samples of freshly stranded oil, and any interpretations of data must take into account alterations in chemical composition due to weathering. The oil in our studies had probably been at sea for at least a week, and had probably lost a large proportion of its lighter smaller molecular weight aliphatics and other components. In this respect the GC trace of oiled loon liver is interesting in that a relatively full spectrum of aliphatics above C₁₂ or C₁₃ were obtained, suggesting that this bird may have become exposed to relatively unweathered original cargo oil.

The question of toxicity is interesting in that toxicity is not an absolute property of the oil but varies directly with weathering of the oil. This is particularly well demonstrated in the results of the laboratory weathering experiment (*viz.* Fig. 6) where both a shift of the curve and a steepening of the curve slope were observed as the sample weathered, suggesting a change from a crude oil to a Bunker C type oil. This aspect of oil toxicity is little studied, but results from long-term field studies in Chedabucto Bay, Nova Scotia, site of the 70000 Bunker C spill in 1970 (*×15* Anonymous, 1970), suggest that long-term residual Bunker C derived hydrocarbons have a persistent and significant toxic effect on metabolism of a number of intertidal infauna (e.g. Thomas, 1978; Gilfillan and Vandermeulen, 1978). This question of changing toxicity with weathering comes up again when considering the potential impact of buried *Amoco Cadiz* oil upon its release from beach sediments during winter beach erosion. For the oil released later will likely not be the same oil buried during the spring depositional phase, having experienced presumably microbial degradation and other weathering processes.

Finally, there is the question of biological response to spilled oil exposure during the initial phases of a major spill. Probably the most striking observation made was the unexpectedly high variability in susceptibility to oil, with extreme mortalities in some species and high survival in others. It would appear that two factors may be involved in this variable impact, one environmental and the other physiological. Near total mortality was observed in marine invertebrate species living freely within the water column, as amongst the crabs, isopods, amphipods and various molluscs. Discounting obvious exceptions there was high survival among benthic infauna, as annelids, nemertean, and

Mya arenaria. (The large mortalities of *Echinocardium* sp., *Enis* sp., *Cardium* sp. and *Venus* sp. may be due to an unexpectedly high and continuously high concentration of hydrocarbons observed throughout the entire subtidal water column reported by Marchand *et al.* [1978], *viz.* these proceedings.) Thus the suggestion arises that intertidal infauna were in part protected by their burrowing habit, whereas the epibenthic organisms live in continuous and total exposure to their seawater environment. Even in the severely oiled marsh sediments of the Ile Grande salt marsh a surprisingly large number of burrowing organisms were found alive below the sediment surface, as was the case in a number of oiled tidal flats.

The second factor, the role of aryl hydrocarbon hydroxylase in tissue detoxification, is equally poorly understood. It is interesting to speculate, however, that the survival of mullet in the awesomely oiled salt marsh channels near Ile Grande may in part have been due to their ability, through induction of the AHH system, to successfully reduce or eliminate petroleum hydrocarbons from their tissues. Evidence for this mechanism of tissue depuration is now accumulating from studies with flounder (McCain *et al.*, 1978) and other systems (e.g. Kurelec *et al.*, 1977). Interestingly, although distribution of this AHH system in the invertebrate phyla is less well understood (Payne, 1977) and in some cases appears nonexistent (bivalves - Lee *et al.*, 1972; Vandermeulen and Penrose, 1978) there is evidence for high tolerance of oiling coupled with a possible role of AHH in certain annelids (Lee *et al.*, 1977; Gordon *et al.*, 1978). This possibility may be a further factor in the observed high survival of many burrowing annelid species.

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