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Contamination of Fish and Shellfish Following Oil Spill Incidents

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ABSTRACT ●

Accidents and spillages are an inevitable consequence of the worldwide transport of crude oil and refined petroleum products by sea. The number of major spills occurring each year has decreased since the 1970s, but spillages and operational discharges from tankers nonetheless constitute a significant input of oil to the marine environment. The rapid uptake of oil and polycyclic aromatic hydrocarbons (PAHs) by exposed fish and shellfish, resulting from large oil spillages in coastal waters, poses a potential threat to human consumers of fish and shellfish and also affects the marketability of catches. In this article, the processes influencing uptake and retention of PAHs in fish and shellfish are outlined by drawing on the experience gained from previous incidents. This includes particular reference to three recent major oil spills from tankers in the United Kingdom and the United States, those involving the Exxon Valdez, Braer, and Sea Empress, and to the resulting contamination of local stocks. A more uniform approach to oil spill monitoring would aid comparisons between spills and would help interpret the kinetics and dynamics of released hydrocarbons in future cases. Currently, no statutory limits or guideline values have been established for PAHs in edible tissues of fish or shellfish, whether derived from oil or combustion sources.

Key Words: crude oil, spills, fish and shellfish, contamination, depuration, tainting, fisheries management, PAHs.

INTRODUCTION ●

Much of the crude oil produced arises in countries such as those of the Middle East, remote from the locations in which it is refined and used. The resulting large-scale and worldwide transport of crude oil as well as the smaller scale and generally more localized transport of refined products generate the potential for tanker accidents and consequent oil spillages along shipping routes and close to ports and harbors. Although tankers are sometimes lost in the open sea, most shipping incidents occur in inshore areas with restricted waters where the potential for damage to fishery resources is greatest (International Petroleum Industry Envi-

ronmental Conservation Association, 1997). The vast majority of spills are small (i.e., <7 tonnes) and arise from routine operations such as loading and discharging of cargo and bunkering operations. Records of these spills are inevitably incomplete, but in most years it is likely that minor spills make a relatively small contribution to the total quantity of oil lost to the marine environment from tankers. Estimates of the annual quantity of oil spilled over the period 1970–1996 range from 9,000 to 608,000 tonnes, with a few very large spills being responsible for a high percentage of the oil spilled (International Tanker Owner's Pollution Federation, 1997). In the 10-year period 1987–1996, for example, there were 362 spills of >7 tonnes of oil, totalling 1372 thousand tonnes, but 992 thousand tonnes were spilled in only ten incidents.

Large spills of oil in coastal waters pose a potential threat to human consumers of fish and shellfish and to the marketability of catches due to the rapid uptake of oil, and therefore polycyclic aromatic hydrocarbons (PAHs), by exposed stocks, both farmed and feral. In this article, the processes affecting uptake and retention of PAHs by fish and shellfish and their consequences for fisheries management and consumers are outlined.

UPTAKE, METABOLISM, AND DEPURATION

Uptake

Marine organisms can be exposed to oil spills ranging in extent from localized events that persist only for days or weeks to major incidents in which oil can remain available for uptake for periods of months or even years (e.g., Teal et al., 1992). In some spills, both situations can be observed in different locations, where both exposed rocky coasts and saltmarshes are oiled. The uptake of PAHs by biota proceeds via two major pathways, directly from water and through the diet (Mackay, 1991; McKim, 1994), with uptake by the former route being more rapid (e.g., Kennedy and Law, 1990; Namdari and Law, 1996). Uptake from water is referred to as bioconcentration and has been observed at many trophic levels both in the laboratory and in the field. Aqueous uptake can be considered as an equilibrium-partitioning process, in which concentrations in water and in an organism's tissues remain in thermodynamic equilibrium.

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Uptake via the diet and so through the food chain is referred to as biomagnification. This process is important only in the lower levels of the food chain for PAHs, because vertebrates have an effective means of biotransforming these contaminants after ingestion (Hellou et al., 1995).

The physicochemical properties of individual PAHs, particularly their degree of lipophilicity, affect their uptake by biota. The octanol-water partition coefficient (Kow) for a compound reflects its water solubility—the larger the Kow value, the more hydrophobic (and more lipophilic) the compound. An increase in the number of rings in PAHs and/or in the level of substitution of a ring will generally decrease the water solubility and so increase lipid affinity (Mackay, 1991). Compounds with a log $K_{ow} < 4$ will be preferentially accumulated from water, whereas those with a log $K_{ow} > 6$ will be taken up primarily from food. Compounds with intermediate values can be accumulated from both food and water (McKim, 1994). Studies of bioaccumulation of hydrocarbons by marine invertebrates have demonstrated that low molecular weight (MW) PAHs (i.e., 2- and 3-ring PAHs), which tend to be more abundant in crude oil, are more water soluble and more abundant in animal tissues than are high MW PAHs (4- and 5-ring PAHs) (Varanasi et al., 1985; Farrington, 1991; Yunker et al., 1995; Hellou and Warren, 1997).

Differences in the mode of uptake due to differences in the feeding behavior of various species of bivalves (e.g., particle or filter-feeders, detritivores) have been documented both in the laboratory and in the field, for instance, following the *Exxon Valdez* and *Braer* oil spills (Grahl-Nielsen et al., 1978; Bender and Huggett, 1989; Shigenaka and Henry, 1995; Hall et al., 1996). Observations relating uptake to feeding strategy also have explained differences in the profile of the accumulated PAHs (e.g., McLeese and Burridge, 1987; Hickey et al., 1995; Meador et al., 1995).

Metabolism

The metabolism of PAHs by organisms is affected by the activity of the mixed-function oxygenase (MFO) enzyme system, which occurs in both vertebrates and invertebrates (Stegeman and Hahn, 1994). However, the activity of the MFO system is generally markedly lower in invertebrates than in vertebrates, and thus a higher level of contaminants is needed to induce MFO enzymes in invertebrates (Fossi et al., 1986; Marsh et al., 1992). These differences in enzymatic response, level, and reactivity of PAHs lead to the bioaccumulation of low MW PAHs in finfish and biotransformation of higher MW PAHs. Invertebrates accumulate a wider range of PAHs due to their lower ability to metabolize xenobiotic compounds (e.g., Hellou *et al.*, 1995; Neff and Burns, 1996).

The metabolism of PAHs proceeds in two stages. Oxidation products are formed in the first stage, and these compounds further react to form various conjugates such as glu-

coronides, sulphates, or amino acid or glutathione derivatives (Hellou et al., 1989; Hellou and Upshall, 1995). These more polar derivatives can then be eliminated from the organisms through the gall bladder bile (Hellou and Payne, 1987). In vertebrates, these reactive first phase oxidation products also can react with macromolecules (such as DNA) to form adducts that interfere with biochemical processes and can ultimately cause cancer.

Depuration

Depuration occurs when the level of oil in the surrounding water declines (McCarthy and Mackay, 1993; Neff and Burns, 1996). Different depuration rates are seen in vertebrates and invertebrates, and they also differ with the length and type of exposure. Short exposures (of the order of hours) result in clearance after a few hours or days, while continuous exposure over a longer period results in both a higher uptake and a longer depuration period (e.g., Farrington et al., 1982; Niimi and Palazzo, 1986; Pruell et al., 1986; Mackay et al., 1992). Certainly, the release of contaminants from oysters exposed to the *Amoco Cadiz* oil spill was slower for the animals exposed over a longer period (Friocourt et al., 1981). Invertebrates often remain contaminated for much longer periods than do vertebrates. After the Exxon Valdez oil spill, PAHs persisted in mussels longer than in salmon (Neff and Burns, 1996), although recontamination probably also played a role in this instance. Salmon (as vertebrates) have the ability to metabolize PAHs, thereby reducing the level of bioaccumulation (Carls et al., 1996), although laboratory experiments with Atlantic salmon also demonstrated that depuration of PAHs from adipocytes was slower than from white muscle tissue (Heras et al., 1992), and decreased depuration rates were correlated with concentrations of higher MW PAHs (Zhou and Ackman, 1994).

METHODOLOGY ●

Chemical Analysis

PAHs derived from combustion processes comprise predominantly parent compounds, whereas those found in crude oil also include a wide range of alkyl-substituted homologs. Whereas both of the major analytical approaches (based on high-performance liquid chromatography and gas chromatography [GC]) can be used for the investigation of pyrogenic PAHs, the greater complexity of oil-derived PAHs requires the use of GC/mass spectrometry (MS) (Law and Biscaya, 1994). Complete separation of all the isomers of the parent and alkylated PAHs is still not possible, but the greater resolving power of capillary GC columns combined with the selectivity of detection by MS maximizes the information produced. Data generated following oil spill incidents are seldom directly comparable with those from other spills, because different groups of PAHs are often determined and

different methodologies also may be used. To some extent, this may reflect differing aims, but more uniformity of approach is desirable. A basic list of PAHs to be determined in monitoring studies that allows the assessment of both combustion and oil sources has been proposed elsewhere (J. Klungsøyr, 1998, personal communication). The application of ultraviolet fluorescence spectrometry as a screening technique between the extraction and cleanup stages provides useful information on the level and range of PAHs in samples and allows the use of the more sophisticated and time-consuming (and expensive) chromatographic techniques to be targeted more effectively (Law et al., 1988).

To monitor the variability due to the sampling and analytical procedures, a full analytical quality control protocol should be instituted within the laboratory program to ensure that the methods remain within control (Law and de Boer, 1995). This topic is considered in detail elsewhere (Quevauviller et al., 1995). Participation in proficiency testing schemes and intercomparison exercises in which "blind" samples are distributed is also recommended, and experience has shown that such programs will improve data quality (Wells et al., 1997).

The number and type of PAHs determined can enable more detailed interpretation of data. In areas of chronic oil contamination, where other spills have occurred, or where there are significant natural oil inputs, it cannot be assumed that contamination must result from a recent spill, and sophisticated chemical fingerprinting techniques may be needed to differentiate contamination from other sources (Brown et al., 1997; Page et al., 1997). Perhaps surprisingly, it is difficult to compare data from different spills to make use of the wealth of previous experience gained around the world. There are a number of reasons for this: because oil spill datasets are often large, they are seldom (if ever) available in full; different suites of PAHs are analyzed; and concentrations may be expressed on either a dry or wet weight basis.

Taint Assessment

The assessment of taint in local seafood is often an important component of oil spill response, particularly where there are extensive mariculture enterprises in coastal waters. Knowledge of the identity of the hydrocarbons that cause taint and the concentrations at which they do so whether alone or collectively is poor; thus, taint must be assessed by sensory rather than analytical procedures. The protocol for taint assessment devised at the Torry Research Station in the United Kingdom (documented by Scottish Office Agriculture, Environment and Fisheries Department (SOAEFD), Aberdeen, in 1996) has been validated by laboratory experiments in which the uptake and depuration of oil taint from fish were studied (Martinsen et al., 1992; Davis et al., 1995). In another series of experiments, the tainting threshold for trout exposed to diesel fuel in ambient water was found to be 0.08 ng/L (Davis et al., 1992), whereas during

oil spills concentrations in the range 1–100 ng/L are commonly encountered, and higher concentrations (>1 mg/L) can result when the rate of dispersion (whether natural or chemically enhanced) is high. These protocols have been used in studies following a large number of spills in the United Kingdom (including those involving the tankers *Braer* and *Sea Empress*) and have proved robust and effective. Although it should be noted that the establishment, training, and maintenance of an effective taint-testing panel can be costly, very large numbers of samples can be examined by such a panel (in the case of the *Braer*, 12,800 individual assessments were made for salmon alone; Whittle et al., 1997). There is often an implicit assumption that an absence of taint is the norm, but this may not be the case in coastal areas with significant inputs from oil facilities or other industries.

Sampling of Fish and Shellfish for Analysis

In the initial stages of an oil spill, the primary aim is to establish the degree and spatial extent of contamination, whilst in the latter stages it is usually to follow the processes of elimination and to chart the return to background concentrations. Ideally, all the important commercial species should be sampled, but if resources are limited then a selection can be made, taking care to consider all the possible routes of exposure and uptake. If samples can be transferred to the laboratory within a short time period (24–48 hr maximum), then they can be shipped on ice, but otherwise they should be frozen before despatch and delivered frozen. When commercial fishing is interrupted, as in the case of a closure, then it may be necessary to charter boats specifically for sampling. The number and frequency of samples to be taken will be a function of the extent of contamination and the analytical resource available, as the flow of analytical data should be sufficiently rapid to feed effectively into the decision-making process regarding fisheries management. Uptake of PAHs can be extremely rapid, particularly in bivalves, whilst depuration is usually a slower process. Following the Sea Empress spill, samples of cockles and mussels were taken from the major commercial beds on a weekly basis for analysis by GC/MS, and this was adequate to chart the elimination of PAHs from their tissues (Law et al., 1997).

FISHERY CLOSURES FOLLOWING OIL SPILLS

The majority of the world's fishing catch comes from the highly productive waters overlying the continental shelves, relatively close to land. From the preceding sections, it is apparent that in the event of a large oil spill in coastal waters then contamination of fish and shellfish stocks is likely. Most likely to be affected are fish and shellfish that cannot evade the oil, and so contamination is likely to be particularly serious in areas with extensive inshore shellfisheries or mariculture (International Petroleum Industry Environmen-

tal Conservation Association, 1991). Farmed stocks were widely contaminated, for instance, in Shetland in 1993 (the Braer; caged salmon [Whittle et al., 1997]), in Spain in 1992 (the Aegean Sea; farmed mussels, oysters, scallops, salmon, and turbot [Hermida-Ameijeiras et al., 1994; Alvarez-Piñeiro et al., 1996]), in Alaska in 1989 (the Exxon Valdez; salmon fry released from hatcheries [Carls et al., 1996]), in France in 1978 (the Amoco Cadiz; farmed mussels and oysters [Berthou et al., 1987]), and in many incidents along the coasts of China, Japan, and South Korea (Moller et al., 1989), where areas of extensive mariculture and heavy shipping activity occur side by side (International Tanker Owner's Pollution Federation, 1996). Damage to seafood on a scale similar to that experienced after the 225,000-tonne spill from the Amoco Cadiz in France has been recorded after spills of 50 tonnes or less in Japan and Korea due to their extensive mariculture (Moller et al., 1989).

Wild stocks also may become contaminated, although fish usually to a lesser extent than crustaceans or mollusks. Following the Exxon Valdez spill in Alaska, however, contamination was found in pollock (Pollachius virens) one year after the spill and >400 miles away (Collier et al., 1993). Pollock are bathypelagic fish feeding in the water column and are a major fishery resource in Alaskan waters. There is no evidence that any oil spill has significantly affected adult fish populations in the open sea, and even where larvae may have suffered extensive mortalities this has seldom been detected subsequently in adult populations. Following the *Amoco Cadiz* spill, however, the 1978 yearclass of plaice (Pleuronectes platessa) and sole (Solea vulgaris) disappeared from the worst affected areas of North Brittany (Centre National pour l'Exploitation des Oceans, 1981). In addition, oysters remained contaminated and their fishery closed for seven years due to oil buried in anaerobic subtidal sediments within the Abers (inlets) which was periodically remobilized by storms and high tides (Berthou et al., 1987).

Once fish and shellfish stocks are (or are likely to be) contaminated, action is usually taken to restrict fishing activity in the affected area. Within the United Kingdom, this is by establishment of a closure area under the Food and Environment Protection Act (1985). The reasons for restricting fishing activity are twofold: the primary aim is to protect human consumers from the risk of eating contaminated food; a secondary (sometimes unstated) aim is the protection of fishery markets. Concern for human health is primarily for the oil-derived carcinogens (some of the high MW PAHs) which may be accumulated, albeit at low concentrations relative to the alkylated derivatives of the low MW PAHs from crude oil. However, epidemiological studies have not demonstrated any risk of cancer or other diseases in humans through eating seafood from areas where oil spills have occurred (GESAMP, 1993). Following the Exxon Valdez oil

spill, the question of whether oil-exposed fish are safe for human consumption was considered with respect to subsistence fisheries in Alaska. The findings of this study were that consumption of flesh from exposed fish involved little risk due to the extensive metabolism and excretion of PAHs by fish, and this finding of minimal risk was supported by an advisory position of the U.S. Food and Drug Administration (Collier et al., 1996). The Food and Drug Administration concluded that the risk of contracting cancer from eating finfish collected from the spill area was so low that it could not be calculated and was therefore for practical purposes close to zero (Bolger et al., 1996). It was also concluded that the risk of contracting cancer from a lifetime of eating shellfish from even the most heavily impacted site was extremely low. These assessments may not be valid in the case of shellfish contaminated by combustion-derived PAHs rather than oil, because the levels of the carcinogenic PAHs may be greater from this source.

Reference to three recent large-scale incidents in the United Kingdom and the United States serves to illustrate some of the problems encountered when fish and shellfish are contaminated following oil spills. ΣPAH refers to the sum of the individual PAH and groups determined in each case.

Exxon Valdez

The Exxon Valdez spilled 38,000 tonnes of Alaskan North Slope crude oil into Prince William Sound in Alaska after running aground on 24 March 1989. Fisheries are critical to the economy and subsistence of residents of the local area (Spies et al., 1996), and the rapid closure of commercial fisheries by the State of Alaska led to concern on the part of native communities and other subsistence seafood users (Mearns and Yender, 1997).

Although most seafood was shown to be uncontaminated by PAHs and untainted, local consumers proved very reluctant to accept these results (Fall and Field, 1996). Fisheries for black cod (sablefish [Anoplopoma fimbria]) and shrimp were closed within the first few days of the spill, mainly because retrieving longlines and pots through the surface of the slick would have contaminated the surfaces of fish and shrimp (Spies et al., 1996). Salmon and herring fishery closures continued throughout the rest of 1989 because shoreline cleanup activities resuspended oil in the nearshore areas where these fisheries were conducted, and fouling of both gear and products was likely. In 1989, PAH concentrations were significantly greater in pink salmon tissues from oiled areas than from nonoiled areas, although by only a factor of three (Σ PAH concentrations 181 \pm 28 versus 54 \pm 8 μ g kg⁻¹ dry weight). In 1990, there was no difference in tissue PAH concentrations between oiled and nonoiled areas (Carls et al., 1996; Kocan et al., 1996; Marty et al., 1997). Dense beds of mussels (Mytilus trossulus) left untreated still showed high levels of contamination (up to 8000 µg kg⁻¹

dry weight) three and four years after the spill (Babcock et al., 1996; Harris et al., 1996). Mean Σ PAH concentrations in these mussels had increased to >100,000 μ g kg $^{-1}$ dry weight within six weeks of the spill where oiling was heavy (Short and Babcock, 1996). Almost all of the beds showing little reduction in contamination were in protected, lowenergy areas, which were not vulnerable to natural disturbances. Therefore, there had been no natural weathering and flushing of the residual oil underlying the mussel beds, and this provided a continuing source of hydrocarbons for uptake by mussels.

Braer

The Braer ran aground in high winds, and stormy weather persisted throughout the spill period. Dispersal of the oil by spraying of chemical dispersants was limited by the strong winds, but \sim 50% of the cargo of 84,700 tonnes of Gullfaks crude oil was dispersed naturally into the water column after release (Davies et al., 1997). The Shetland seafood industry dominates the economy of the Islands and is vital to their long-term future, making the Shetland Islands one of the most fisheries-dependent regions within Europe (Goodlad, 1996). A fishery closure order was implemented three days after the grounding of the Braer to prevent contaminated seafood reaching the market. Rapid uptake of PAHs by salmon caged in farms on the west of Shetland resulted in concentrations of Σ PAH in muscle tissue up to 14,000 µg kg^{-1} wet weight (compared to a mean background ΣPAH concentration of 30 µg kg⁻¹) and led to the destruction of 50% of the 1991 intake of smolts (due to be harvested during 1993) and the entire 1992 intake. The maximum concentrations of PAHs in wild finfish muscle was 2650 µg kg⁻¹ (in whiting sampled in January 1993). These concentrations declined rapidly, and the exclusion zone for wild fish was lifted within four months of the grounding (Topping et al., 1997). Other restrictions were lifted in September 1994 (for crustaceans other than Nephrops norvegicus) and February 1995 (scallops [Pecten maximus]). However, the taking of mussels and Nephrops from some sites in the closure area is still prohibited over six years after the spill as a result of oil-contaminated sediments in the upper voes (inlets) in the area (similar to the situation on the north coast of Brittany following the *Amoco Cadiz* spill in 1978).

Sea Empress

The tanker *Sea Empress* grounded near the mouth of Milford Haven in southwest Wales on 15 February 1996 and spilled ~72,000 tonnes of Forties blend crude oil and 480 tonnes of heavy fuel oil (Sea Empress Environmental Evaluation Committee, 1998). The waters off south and west Wales support a diverse fishery for both fish and shellfish, and a fishery closure area was implemented (Law et al., 1997, 1998). These restrictions were removed in stages by species

and area. Fish took up only low concentrations of low MW PAHs (naphthalenes and phenanthrenes) which were rapidly lost, and restrictions covering finfish were removed within approximately three months of the grounding. The highest ΣPAH concentration seen in fish muscle was 259 μg kg⁻¹ wet weight in a sample of bass, but most values were <100 μg kg⁻¹. Other than within Milford Haven itself, PAH concentrations in crustaceans also remained low, and as in the case of finfish, low MW PAH dominated. Mollusks (and mussels in particular) accumulated much higher concentrations of PAH than did fish or crustaceans. At sites ~4 km from the spill site, mussel tissue contained 6800 mg kg⁻¹ of oil and $>100,000 \mu g kg^{-1}$ wet weight of ΣPAH (mostly alkylated naphthalenes and phenanthrenes) within nine days of the grounding. Once the release of oil had ceased, these concentrations began to decline, and the final restrictions (applying mainly to intertidal mussels, which retained contamination the longest) were lifted on 12 September 1997, \sim 19 months after the grounding.

REMOVAL OF FISHERY RESTRICTIONS

Once fisheries have been closed, the criteria for lifting restrictions need to be clearly established. In the cases of the *Braer* and the *Sea Empress*, in which closures were enacted under the same legislation although for different countries of the United Kingdom, similar criteria were established:

- concentrations of hydrocarbons and PAHs in fish and shellfish tissues within the range of background values established for reference sites outside the exclusion zone or for similar samples collected within the zone but before the spill; and
- an absence of taint associated with crude oils and petroleum fractions in tissues of fish and shellfish sampled within the zone.

Background concentrations of PAHs in fish and shellfish will never be zero if appropriate analytical techniques are employed; PAHs are produced by both natural and anthropogenic processes and are ubiquitous environmental contaminants, and oil is released from natural seeps (Fileman and Law, 1988; Volkman et al., 1992; Law and Biscaya, 1994; Hellou and Warren, 1997; Page et al., 1997). Background data collected before a spill are seldom if ever available for the full range of species and locations affected and may be entirely absent. Data gathered within programs mounted for general environmental surveillance, such as the National Status and Trends program in the United States, can be of use in the event of a spill. However, these samples are collected annually at the same time each year, and thus the data do not reflect the seasonal cycles in tissue concentrations of PAHs in mollusks (Jacob et al., 1997; C. A. Kelly, 1998, personal communication).

Commercially exploited shellfish beds and mariculture facilities may be located in estuaries and coastal waters that are contaminated as a result of local inputs from urban and industrial sources, the latter often combustion derived. However, there are generally no statutory limits or guideline values established for PAHs in fish and shellfish tissues against which to assess contamination in relation to consumption and sale. Establishment of such values would not be straightforward and may vary from country to country, being most stringent in countries in which seafood forms the highest proportion of the local diet. What action should be taken therefore if tainting or contamination persist but it is related to sources other than the original oil spill? One argument states that these levels of contamination preceded the spill and that utilization of the resources was sanctioned then; thus, the fisheries should be reopened. The second argument says that the closure should be maintained as it would have been if the spill were responsible. This is likely to be an indefinite closure and may be difficult to justify in the absence of statutory limits or guideline values, particularly as people may be put out of work as a result. It may also result in an anomalous situation, whereby seafood with higher levels of PAHs than those persisting from the spillaffected area can be caught in other areas and sold.

CONCLUSIONS •

A more uniform approach to oil spill monitoring and a greater comparability of methods and determinants studied would aid comparisons between spills, and lessons could be more easily learned than at present. A scientific understanding of the processes underlying the uptake of PAHs by fish and shellfish informs the decision makers on fishery management issues, but it should not be forgotten that ultimately these decisions are political as well as scientific. This is always a complex issue, and public and media concern, consumer and market confidence, and the representations of fishermen will always be factors to be taken into account at the political level. Studies undertaken following the Exxon *Valdez* oil spill suggest that there is little risk to consumers from eating fish and shellfish contaminated with oil; however, no statutory limits or guideline values have been established for PAHs in edible tissues, whether derived from oil or combustion sources.

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