

Flavors in Fish From Petroleum Pickup

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ABSTRACT — All flavors noted in fish resembling petroleum oil are not derived from oil in water. Origins of various flavors found in fish are discussed. Many facts needed to understand the relationships between levels of petroleum in water and in fish and flavor of the fish have not been elucidated. Particularly, there are large discrepancies between reports by different investigators regarding levels of hydrocarbons and resulting flavor intensities. More research is needed to resolve these contradictions.

INTRODUCTION

It has been common knowledge for many years that when fish are caught and stored aboard the fishing vessel in ice, and bilge water containing oil accidentally comes in contact with the fish, the quality of the fish is impaired from a resulting off-flavor development in the flesh. The off-flavor can range from only a slight impairment, in cases where only small amounts of oil are involved, to a condition where the fish are completely ruined for human consumption. This kind of accidental contamination of fish has occurred frequently enough that there can be no doubt that when fish are stored with access to sufficient petroleum oil, a damaging alteration in flavor, which has generally been designated as a taint, develops.

In such cases as have just been described, the fish are dead at the time they have been in contact with water containing petroleum. We cannot justifiably conclude from such incidents that live fish living within waters contaminated by petroleum would develop such a taint. It appears quite likely that in the case of live fish, at least a part, if not all, of any petroleum entering the fish would, by the processes of metabolism, be altered, perhaps excreted so that there might be little or no alteration in the fish's flavor. It is the purpose of this paper to consider research findings dealing with the pick-up of petroleum by fish living in

petroleum-contaminated waters with reference to alteration in flavor of their flesh.

Only a relatively small amount of research has been carried out on such matters as the effect of petroleum upon flavor of fish living in a petroleum contaminated environment. This is partly because interest in such environmental problems has not, until quite recently, been evident. A further hindrance to such research is caused by the rather difficult nature of such studies. Flavor of fish, or of any food material, is a completely subjective measurement which does not readily lend itself to use of scientific techniques. For direct measurement of flavors, it is necessary to use highly trained flavor panels and even when this is done, the subjective nature of the tests often leaves questions as to the accuracy of the results. On the other hand, it is possible to study flavors in an indirect manner by chemically measuring the components responsible for the different flavors. In the present instance, for example, a comparison of the content of hydrocarbons known to occur in petroleum found in fish living in petroleum-contaminated water would give presumptive evidence, at

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least to the effect that there might be flavor problems.

Before discussion of research which has been carried out on matters specific to our interest, a short introductory section is included to give background on different types of flavors occurring in fish and from what sources they are derived. This information is needed in order to understand the differentiation between baseline flavor of fish in the absence of petroleum from flavors directly connected with the presence of petroleum.

GENERAL NATURE OF FLAVORS AND ODORS IN FISH

Three types of flavors, based upon the source from which they are derived, occur in fish, and each of these will be described briefly. These are natural flavors, unnatural flavors, and decomposition flavors.

Each species of fish contains many different flavor chemical components which blend together to give that species a particular flavor. Just as such other flesh foods as beef, pork, lamb, and chicken have composite flavors which enable us to distinguish one from another, so also such species as salmon, herring, and cod have different natural flavors, each characteristic for the different species. The flavoring components characteristic of each species occur both in the oil and in the protein of the fish. Ordinarily the most flavorful components are chemical entities which are dissolved in the oil of the fish. Nevertheless, fish protein also possesses some characteristic, though often quite bland, flavors.

Unnatural flavors occur in occasional specimens of a species which give these fish flavors not typical for the species. Such flavors occur especially when fish are consuming some type of feed which they normally do not eat. This may give rise to abnormal flavors, which sometimes are quite pronounced and which may be undesirable. For example, during certain seasons flatfish, such as sole, feed upon certain plankton which give rise to a flavor reminiscent of that of iodine. In other cases fish may consume, during a small portion of their

life span, feed giving a wide variety of abnormal flavors. In some cases, such flavors might resemble that of petroleum, even though no petroleum was involved. In other cases, the fish may acquire unnatural flavors by coming in contact with substances accidentally introduced into the water, petroleum being an example of this type of source. The fish could acquire such material by ingesting it through the digestive tract, by taking it into their respiratory system through the gills, or perhaps in some cases by direct absorption through the skin.

Decomposition of fish is the third process which can lead to off odors and flavors in fish. Bacterial decomposition (spoilage) leads to production of mostly volatile components like trimethylamine, ammonia, and hydrogen sulfide. These result in off odors, but during cooking of the fish most of the compounds responsible for the odors are volatilized so that the flavor of the cooked fish is altered much less than might have been indicated from odor observations. The components responsible for the natural flavors characteristic of the various species are highly labile and such flavors disappear rapidly, due probably to oxidation, as the fish are held at refrigerated temperatures.

Lipids (mostly oils) of fish are oxidized to some extent during storage in ice and to an extensive degree for fish frozen and held in cold storage. The oxidation products, which are mostly carbonyls, give rise to rancid types of flavors.

For research on flavors in fish the most common approach used by scientists has been to attempt to isolate from the fish chemical constituents most probably responsible for the flavor. Such research is time consuming and it involves intricate separation procedures to isolate the chemical constituents making up the flavor from hundreds or thousands of other compounds which are also present. The flavor components occur ordinarily in only trace amounts, which makes it most difficult to separate them from the other components, many of which are present in amounts several orders of magnitude greater than the flavor components. Identification of the isolated

components is usually confirmed by mass spectrometry, sometimes used in conjunction with other criteria.

Many investigations do not go beyond the stage of isolation and identification of components which theory would indicate are likely causes of the flavor. In order to give the research results any real meaning, it is necessary to confirm that the chemical components isolated and identified really do possess the flavor of the fish which is noticed when the cooked fish is eaten. In order to carry out this additional research it is necessary to use a highly trained sensory evaluation panel of experts who are thoroughly familiar with the different flavors associated with fish. Such a panel can positively confirm whether the flavor of the fish is caused by the mixture of isolated and identified components and also can establish the ranges of concentration of the different components which make up the flavor. Unless this final step of a flavor evaluation is carried out, the results of a purely chemical study may be of little value from any practical application standpoint.

RESEARCH ON FLAVORS PICKED UP FROM PETROLEUM

The number of investigations and papers in this field is not huge and since most of the articles that have appeared deal with some limited aspect such as analytical methods, or are of a very general nature, the number of research projects set up to learn the nature of the phenomena involved or looking at the overall picture are very few indeed. Most of this review will deal with these latter types of research projects.

Some of the most comprehensive research in this field is coming from Torry Research Station (Mackie et al., 1972; Hardy et al., 1974; Howgate et al., 1976). These investigations include, for example, research on hydrocarbons isolated from brown trout caught in a stream which had been polluted by a 2,000 gallon diesel oil spill. The hydrocarbons were separated chemically from other constituents of the fish and were shown to be identical to hydrocarbons contained in the diesel oil. Furthermore, the odors and flavors of the hydrocarbons isolated from the

fish matched exactly those from the hydrocarbons from samples from the same batches of diesel oil as had been spilled in the stream. Hydrocarbons in small trace amounts were isolated from brown trout caught in the same stream at sufficient distance upstream from the site of the spill so that no petroleum was present in the water. The odor of these hydrocarbons had no similarity to those from the polluted fish and were doubtlessly of biogenic origin. The odor and flavor of the hydrocarbons from these control fish were almost nil and were not at all similar to that from the hydrocarbons from the oil polluted fish. In another set of experiments by Torry Research Laboratory investigators (Hardy et al., 1974) the rate of uptake of hydrocarbons into the liver of wild codlings was measured. The fish were kept in tanks of seawater at the laboratory and the fish were fed cod liver oil containing Kuwait crude petroleum. As controls in another batch of fish, the oil fed was entirely cod liver oil. In these experiments there was a preferential absorption of carbon chain length of around 26 from the fish fed the petroleum but not from the controls. Hydrocarbons of chain length C_{15} to C_{21} were absorbed to an insignificant extent. In the hydrocarbon chain length range from C_{22} to C_{26} there was an increasing amount absorbed up to a maximum at chain length C_{26} . From chain length C_{26} to C_{30} there was a decreasing extent of absorption down to chain length C_{31} , at which no significant absorption into the codling livers took place. There was in no instance any evidence of preferential absorption by odd as contrasted to even hydrocarbon chain lengths. The first preliminary experiment, while too limited in scope to be conclusive, indicates that the codling possesses ability to preferentially absorb hydrocarbons of certain chain length. During a 6-month period after cessation of feeding of the crude petroleum by the fish, hydrocarbon levels had fallen to about half the maximum levels reached but were still about 65 times higher than in the control samples.

In another investigation, scientists in Australia's CSIRO (Commonwealth Scientific and Industrial Research Organization) Division of Food Preserva-

tion have been approaching this problem in a somewhat different way. For example, Shipton et al. (1970) investigated a kerosene-like flavor in mullet. Upon solvent extraction of such fish and examination and separation of hydrocarbons, they found characteristics similar to those of hydrocarbons found in crude oil. In other cases, however, Vale et al. (1970) found compounds apparently responsible for the kerosene-like flavors which arose from thermal decomposition of naturally occurring components in fish.

Deshimaru (1971) studied the effect on fish flavor of rearing yellowtail, *Seriola dorsalis*, in water containing crude petroleum oil. The amount of the crude oil in the fish rearing tanks was set at two levels, 10 ppm and 50 ppm, in different portions of the experiment. A third group of fish was held in water containing no petroleum but was fed feed consisting of fish containing 1 percent crude petroleum oil. The meat from the fish reared with 50 ppm oil in the water had a strong fishy flavor if the fish had been exposed to the oil-containing water for at least 5 days. No fish odor was observed, however, even after 13 days rearing with the oil when the level of oil in the water was only 10 ppm. A fishy flavor developed after more than 13 days exposure, but only to a slight degree. When the fish were fed with oil at the 1 percent level in the feed, a fishy flavor developed at a very low level after 5 days under test, but the degree of intensity had not increased by the end of the experiment (13 days). The presence of a C₂₇ paraffin hydrocarbon was identified by gas chromatograph analysis of the flesh of yellowtail for the samples that had been held in water containing both 50 ppm and 10 ppm levels of petroleum.

Wilder¹ carried out quite different types of experiments on lobster meat and lobster liver (tomalley). In these experiments the living lobsters were coated with a fairly heavy layer of Bunker C petroleum oil. The lobsters were then, in some cases, returned to seawater in a 48-gallon tank in which

the water was replaced at the rate of 1.6 gallons per minute. After exposure to running seawater in this way, lobsters were observed for disappearance of the oil. Within 6 hours less than 10 percent of the oil remained on the lobsters. After 96 hours only a few droplets of oil were visible at crevices on the ventral surface. Lobsters were then cooked and both the flesh and the liver examined by a taste panel. After 192 hours, more lobsters were withdrawn and subjected to the same treatment. In neither the meat nor the liver, after either 4 or 8 days exposure to the running seawater, was there any evidence of taint found by the panel. In another part of the experiment lobsters were fed bait covered with Bunker C oil. The lobsters readily consumed the bait, but again there was no evidence of taint after 4 or 8 days exposure to running seawater.

On the other hand, when untreated lobster were held in a tank of seawater containing 1 part of Bunker C oil per 1,000 parts of water (1,000 ppm) for 90 hours, both the meat and liver of the lobsters after cooking had a very decidedly objectionable oily flavor. The liver samples had a stronger flavor than did the lobster meat. When the lobster treated in this way were held live in running seawater for as long as 3 weeks, the oily taste in the meat and liver, after the lobsters were cooked, still remained although at reduced intensity.

Some additional tests indicated that oil could be more completely removed from lobster by wiping if the dispersant, Corexit,² is used in water employed for the deoiling process.

A classical example of a petroleum-like odor in fish, which upon investigation proved to be something quite unrelated to petroleum, was documented by Motohiro (1962). As much as 5 percent of the canned chum salmon produced by the Japanese in the Bering Sea possessed a distinctly petroleum-like odor. After an extensive investigation, it was found that the odor was from dimethylsulfide which came from the precursor, dimethyl-2-carboxyethyl sulfonium sulfide. This substance was found in the

digestive tract of a portion of the chum salmon packed in the north Pacific and it is believed to arise from certain algae consumed on occasion by salmon. Although the odor of the fish had what most observers described as a petroleum-type odor, it was shown conclusively that no petroleum product was involved in formation of the odor.

Mullet, possessing a petroleum type of flavor, have in some cases been found among the Australian catch. In a preliminary investigation, Grant (1969) suggested that, like the petroleum flavor found in canned chum salmon, this flavor in mullet might stem from decomposition of feed components to produce dimethylsulfide. In more extensive investigations, Connell (1971, 1974) showed that this mullet off-flavor arose from petroleum contamination. The volatile constituents from the tainted mullet were steam distilled and examined by gas chromatography-mass spectrometry. The resulting gas chromatograms and mass spectra showed close resemblance to those from commercial kerosene. It was also found that sediments in the Brisbane River in which the mullet spent part of their time also contained substances having gas chromatography-mass spectrometry characteristics similar to kerosene. It was concluded that the mullet probably included some of these sediments with their feed although they may have picked it up from the water.

Nitta et al. (1965) investigated the disagreeable odors found in fish occurring near petroleum refining plants at Yokkaichi, Japan. It was found that the odor of the fish flesh was similar to the odor of the effluent from the petroleum refineries. The investigation revealed that the odor could be picked up either from the water in the effluent or from bottom muds near the refinery. The concentrations of ether extract substances which were able to produce this effect were 0.01 ppm in water or 0.2 percent in the muds. It was determined that the substances causing the odors entered the fish through the respiratory system (gills). The odor became apparent in the fish flesh after only about 24 hours exposure to the effluents. Fish did not avoid the oily effluents; on the contrary, the fish seemed to be attracted by them.

¹Wilder, D. G. 1970. The tainting of lobster meat by Bunker C oil alone or in combination with the dispersant, Corexit. Unpubl. manuscript, 23 p. Fish. Res. Board Can. Biol. Stn., St. Andrews, N.B.

²Reference to trade names does not imply endorsement by the National Marine Fisheries Service, NOAA.

Other investigations correlating petroleum-derived flavor or odor of fish with petroleum refining operations have been carried out (Krishnaswami and Kupchanko, 1969; Miyake, 1967; Ogata and Miyake, 1970; Yoshida and Uezumi, 1961; Yoshida et al., 1967). Of particular interest here, however, is the relationship between offshore drilling and oil taint (Mackin and Sparks, 1962; St. Amant, 1958; Menzel^{3, 4}). The State of Louisiana (St. Amant, 1958) tabulated 60 complaints of oysterermen with operations near offshore oil wells with about 40 percent of the complaints dealing with oily taint. Investigations showed that mud at oyster beds near such drilling operations had very high ether soluble extracts (up to 14,000 ppm). Usually the content of such extractives decreased as one moved away from the vicinity of the drilling operation, but in a few instances maximum oil contamination of the mud was at a distance of 500 feet or more from the site of the wells. It was found that this situation usually occurred with wells deeper than 10,000 feet and especially where the drilling operation employed a mixture of diesel oil with drilling muds. It was found that such drilling operations could result in tainted oysters if the oyster beds were within 5,000 feet of the wells in closed channels or within 2,500 feet in open bays. The oily taste in such oysters was found to persist for as long as 4-6 months after oil pollution ceased.

Oily taste from petroleum has been reported for shellfish other than oysters. Such reports have dealt with mussels (Nelson-Smith, 1970; Brunies, 1971) and clams (Hawkes, 1961).

GENERAL DISCUSSION

The amount of research carried out on the development of an off flavor, sometimes designated as taint, by absorption of petroleum oil or its components is not at all extensive. Fairly numerous articles have dealt, often

superficially, with the presence of such flavors in fish, but very few research papers have been published in which the subject is investigated, even covering only a small aspect, in any kind of thorough and systematic way. I could find no research results on development of such flavors under Arctic conditions.

Petroleum-like flavors have been widely reported to occur in fish, but frequently when such flavors are further investigated they are found to be derived not from any petroleum precursor but rather from some alteration in naturally occurring components of the fish. Nevertheless enough instances of petroleum-like flavors in fish have been investigated and fully documented that the flavor arose from a petroleum product or one of its components, to leave no doubt that such products definitely can give rise to off flavors in fish.

Taint type flavors can occur by mere contact of the surface of the fish with petroleum. This occurs only when relatively large quantities of petroleum are involved. The petroleum and its flavor can in such cases be completely removed, or nearly so, by washing in running water with the removal facilitated by use of a detergent. In the more usual case the petroleum is picked up from the water by the fish either directly from the water through the respiratory system or from feed through the digestive tract. In such cases much less oil is required to bring about the effect and the off flavor is removed only by removing the fish from a source of oil. The pickup of oil in these cases is usually a slow process requiring days or weeks of exposure and reduction and elimination of the flavor is an even slower process requiring weeks or months of residence of the fish in an oil-free environment for complete removal of flavor or odor.

The vast predominance of research in which fish are exposed to waters containing petroleum or its components and then, after suitable intervals, are examined for flavor pickup have used relatively high concentrations of oil in the water, far more than ordinarily would occur under normal circumstances in the natural environment except under most unusual circumstances. Levels used generally are of the order of parts per million. For

example Deshimaru (1971) exposed fish to water containing either 10 ppm or 50 ppm oil and fed fish feed containing 10,000 ppm of oil; Wilder (see footnote 1) exposed lobsters in tanks of water containing 1,000 ppm oil. The level of oil in waters occurring as a result of discharge of oil by industrial operations are many orders of magnitude lower than such levels. Ordinarily, levels are at parts per billion, even at parts per trillion, concentration. The only cases where many parts per million levels would occur are immediately adjacent to a massive oil spill or perhaps at a discharge pipe coming directly from a processing or manufacturing plant. Even then such levels of the order of parts per million are very transitory and owing to dilution soon diminish to lower levels. Because of these circumstances the results of most of the research reported in the literature on taint-like flavor picked up in controlled experiments does not necessarily reflect what happens in the usual environment. Based upon the degree and rate of development of taint in what controlled experiments as have been reported and where far greater levels of oil were used than would be encountered in open natural waters, it is the belief of this reviewer that taint from petroleum oil would never result from fish living in open ocean waters (barring some major catastrophe such as the *Torrey Canyon* incident, and then only for a short time span). Taint is a common occurrence when fish are accidentally dipped in oil or oil-water emulsions such as might occur in a restricted hold aboard a fishing vessel. It probably also occurs occasionally in streams on very restricted bays or harbors where large volumes of oil accidentally come in contact with the fish. There is no evidence for its occurrence either for fish in the open sea or even in large bays with moderate to considerable discharge of oil from industrial operations.

Information on the level of petroleum-derived components in fish which barely are sufficiently high to cause detectable flavor are still too incomplete to give any indication of what levels might be tolerable. Ineson and Packham (1967) report threshold odor concentrations for certain petroleum products in water. These range around

³Menzel, R. W. 1947. Observations and conclusions on the oily tasting oysters near the tank battery of the Texas Co. in Crooked Bayou at Bay Ste. Elaine. Unpubl. rep., 6 p. Tex. A&M Res. Found., Proj. 9.

⁴Menzel, R. W. 1948. Report on two cases of "oily tasting" oysters at Bay Ste. Elaine oilfield. Unpubl. rep., 9 p. Tex. A&M Res. Found., Proj. 9.

100 ppb to 2 ppm for most crude or refined petroleum oil. Research is needed to yield information on maximum levels of such compounds that can occur in fish without resulting in any perceivable off odor or flavor.

What, then, do we know, based upon the few experiments described? With the limitations that most of the experimental work has dealt with concentration of petroleum several orders of magnitude greater than occurs in open waters and that none of the research was carried out under Arctic conditions, the following conclusions can at least tentatively be reached.

Petroleum reaches the marine organisms primarily through the water column, but may come from heavily contaminated mud or other sedimentary bottom. A major mode of entry is through the respiratory system, although it can also be picked up in the feed and there is no evidence at all to rule out that the food chain may not be of equal importance as a mode of entry. Taint types of flavors have been reported in at least one instance (Nitta et al., 1965) with as little as 0.01 ppm petroleum in water which has been in contact with fish for no longer than 24 hours.

Fish preferentially absorb hydrocarbons with chain length in the range of C₂₂ to C₃₀ with a maximum at C₂₆, and odd and even carbon chain length hydrocarbons are absorbed at about the same rates. There is no agreement among investigators as to the levels of petroleum in water needed to bring about off flavors in the meat of the fish. Thus Deshimaru (1971) reported that fish in waters containing 10 ppm of petroleum within 13 days exposure developed no fishy odor, yet Nitta et al. (1965), in experiments in natural waters, reported definite off odors developed in fish exposed for only 24 hours to levels of petroleum of only 0.01 ppm. While differences in type of petroleum and varieties of fish in different instances doubtlessly are important, such wide differences in threshold concentrations of three orders of magnitude plus 13 times difference in exposure time seem hard to account for on any such basis.

Retention of off odors after removal from the pollution may require as long

as 6 months or more before disappearance of the off flavor. On the other hand, if the contamination occurs only as a result of external application, as in the experiments of Wilder (see footnote 1) the off-flavored meat of the fish can be returned to normal after only a few hours holding in pure running water.

Although several investigators have isolated, from fish or other marine organisms which possessed an off or taint odor, compounds having chemical characteristics closely resembling petroleum hydrocarbons, there has been no identification of single compounds nor groups of mixed, identified compounds which possess the characteristic "taint" flavor resulting from exposure of marine organisms to petroleum.

The area of research in this field which has been most neglected and in which more work should certainly be undertaken is a definitive evaluation of the maximum concentration of different kinds of petroleum in water in which fish can reside without picking up petroleum types of flavors. In what few reports on this matter exist in the literature there is such a complete disagreement among levels found which are said to lead to petroleum flavors or odors in the fish that we really know nothing definite.

A recent research trend has been toward looking at what happens to fish when they live at the bottom near sediments which contain oil. At Torrey Research Laboratory (Howgate et al., 1976), research is at an early stage on looking at effects on flavor of fish which have been kept close to mixtures of sand and petroleum under laboratory holding conditions. Work on more general effects of petroleum on fish held under similar laboratory conditions is underway at the Northwest and Alaska Fisheries Center. Most research up until recently has involved effect of oil suspended or dissolved in the water column. It will be interesting to find what differences occur between effect of oil in sediments and oil in the water column as such research proceeds.

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