Toxic Chemicals, Including Aromatic and Chlorinated Hydrocarbons and Their Derivatives, and Liver Lesions in White Croaker (Genyonemus lineatus) from the Vicinity of Los Angeles

Donald C. Mallins,* Bruce B. McCain, Donald W. Brown, Mark S. Myers, Margaret M. Kranh, and Sin-Lam Chan


High concentrations of toxic chemicals in sediment and white croaker (Genyonemus lineatus), as well as liver diseases (e.g., carcinomas) in this species, were found in the Los Angeles area. The highest concentrations of aromatic hydrocarbons (AHs) in the sediment were in San Pedro Bay, and the highest concentrations of 1,1,1-trichloro-2,2-bis(p-chlorophenyl)ethane (DDT) derivatives were in sediment from near the White Point sewer outfall. Concentrations of AHs, polychlorobiphenyls (PCBs), and DDT derivatives were generally higher in food organisms (benthic invertebrates) from the croaker's stomach than in sediment. Moreover, croaker from San Pedro Bay and White Point were substantially contaminated with DDT derivatives and metabolites of aromatic compounds (in bile), compared to croaker from the Hyperion outfall and Dana Point (reference area). The evidence suggests that the observed pathological conditions of the liver were associated with exposure of the croaker to toxic chemicals, which occurred, at least in part, through the ingestion of contaminated food organisms.

Introduction

Bottom-feeding fish in urban coastal waters are exposed to myriad toxic chemicals (1, 2), and several studies indicate that many of the chemicals accumulate in these fish (3, 4) and thus create a potential for altering their health (5-6). For example, accumulations of metabolites of toxic chemicals in the bile (9, 10), of English sole, Parophrys vetulus, from Puget Sound were recently shown to be associated with liver diseases, including liver cancers. In another study (8), similar associations were observed between toxic chemicals [e.g., aromatic hydrocarbons (AHs)] in sediments and various diseases in English sole.

Marine waters adjacent to Los Angeles are known to receive considerable amounts of industrial and municipal wastes (11-13). This environment thus affords an opportunity to expand limited knowledge available on the bioaccumulation, disposition, and food chain transfer of toxic chemicals. White croaker Genyonemus lineatus, were of particular interest because of their wide distribution along the California coast and because they are an important component of the skiff sports fishery. This bottom-feeding fish also forms the basis for a growing gill net fishery and is a mainstay of pier catches in Southern and Central California (14). Love et al. (14) reported that adult white croaker spawn in shallow waters (5-12 m) and younger fish tend to reside in shallow waters, migrating to deeper waters (22-36 m) as they become mature adults. Accordingly, chemical analyses were performed on sediments and on stomach contents (food organisms), liver, and bile from white croaker collected in the Los Angeles area (San Pedro Bay and near the 5-mi Hyperion and White Point sewer outfalls) and a nonurban reference area (Dana Point) (Figure 1). Observations were also made on histopathologic conditions in these fish.

Methods and Materials

Sediment samples and white croaker were collected in December 1984 from San Pedro Bay (Queen'sway Bay, Cerritos Channel, and near Reservation Point) and from the vicinity of the White Point and 5-mi Hyperion sewer outfalls (Figure 1). Comparable samples were obtained from Dana Point in September 1984. Surface sediments (top 2 cm) were collected with a modified...
at each site by otter trawl, were measured (mm), weighed (g), and necropsied. At each site, the following samples were collected for chemical analyses: bile, from 10 individual fish; stomach contents (food organisms), a composite from 10 fish; liver, a composite from 5 fish. The mean lengths (mm) of fish from each site used for chemical analyses were as follows: Queensway Bay, 221 ± 1; Cerritos Channel, 163 ± 13; Reservation Point, 200 ± 36; White Point, 201 ± 7; Hyperion, 255 ± 7; Dana Point, 191 ± 21. All samples were kept at -20 °C until analyzed.

Sediments and stomach contents were analyzed for a broad spectrum of AHs and chlorinated hydrocarbons (CHs) by using capillary column gas chromatography with mass spectrometry, flame ionization, and electron capture detectors (15). A high-pressure liquid chromatographic/fluorescence detection technique (9,10) was employed to measure metabolites of aromatic compounds in bile. This technique was used because analyses of AHs (e.g., components of fossil fuels and their combustion products) in tissues are of limited value due to the extensive metabolism of these compounds, especially in the liver (16-18). Samples of liver tissues were analyzed (15) for the more metabolically resistant CHs (2). Stomach contents from a composite of five fish were also collected at each site and preserved in 10% neutral, buffered formalin for taxonomic characterization. Also, as part of the fish necropsy procedure, liver tissue was routinely collected for histopathological examination and preserved and processed by previously reported methods (5). Lesion classification followed previously described diagnostic criteria (19-23).

Results

Chemicals in Sediments and Stomach Contents. Sediment-associated AHs, including benzo(a)pyrene (BaP), were found at the San Pedro Bay sites at summed concentrations of 890-2800 ng/g dry weight. ("Summed concentrations" refers to total concentrations of compounds in Table I; all concentrations for sediments, stomach contents, and liver are on a dry-weight basis.) Concentrations of AHs in sediment from Dana Point were generally close to, or below, the limits of detection (Tab. I). The highest concentrations of 1,1,1-trichloro-2,2-bis(p-chlorophenyl)ethane- (DDT-) related compounds and PCBs were found in sediments from the vicinity of the White Point sewer outfalls (summed concentrations, 130 and 520 ng/g, respectively). The concentrations of DDT- and related compounds were 15 and 100 times lower than those previously reported for sediments collected in the vicinity of this site by Brown et al. (24) in 1982 and by Young et al. (25) in 1976, respectively. These differences do not necessarily imply reductions in sediment concentrations over time; they could be due to an uneven distribution of chemical contaminants in surface sediments at the White Point site.

Stomach contents (food organisms) of white croaker captured inside San Pedro Bay contained substantially higher summed concentrations of AHs than did samples from the Hyperion and White Point sites (Table I). For example, the stomach contents from Cerritos Channel had 20 times the summed concentrations of AHs (30000 ng/g) than did the sample from the Hyperion site. Concentrations of AHs in the stomach organisms from Dana Point were all close to or below the limits of analytical detection (Table I).

Highest concentrations of CHs in the stomach contents were found in fish from near White Point; the summed concentrations of DDT-related compounds and PCBs were 13000 and 1000 ng/g, respectively (Table I). DDT and...
Table 1. Aromatic and Chlorinated Compounds (ng/g, Dry Weight) in Sediment (S), Stomach Contents (SC), and Livers (L) of White Croaker from the Los Angeles Area and a Reference Area (Dana Point)*

<table>
<thead>
<tr>
<th>Compoundsa</th>
<th>Queenway Bay (1)</th>
<th>Carrot Channel (2)</th>
<th>Reservation Point (3)</th>
<th>White Point (4)</th>
<th>Hyperion (5)</th>
<th>Dana Point (6)</th>
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<tr>
<td>aromatic hydrocarbons</td>
<td></td>
<td></td>
<td></td>
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<td></td>
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<tr>
<td>methylphenanthrene</td>
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<td>410</td>
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<td>17</td>
<td>120</td>
<td>&lt;4</td>
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<td>1100</td>
<td>&lt;3</td>
<td>&lt;2</td>
<td>190</td>
<td>&lt;11</td>
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<td>520</td>
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<td>&lt;3</td>
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<td>29</td>
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<tr>
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<td>1100</td>
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<tr>
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<td>42</td>
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<td>100</td>
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<tr>
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<tr>
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<td>5</td>
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<td>2400</td>
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<td>6</td>
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<td>1000</td>
<td>170</td>
<td>140</td>
<td>150</td>
<td>1000</td>
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</tbody>
</table>

* Only major components in each category of compounds are presented. a Concentrations were calculated with internal standards (16). b Aromatic hydrocarbons were not measured in liver. c The less than symbol (<) indicates that the chemical was not detected; the value given is the detection limit. d Not applicable.
The mean percentages, by weight, of the identifiable food organisms for the four sites were as follows:

- Queensway Bay: 13% polychaetes, 41% crustaceans, 21% small fish, and 7% nemertean worms
- Cerritos Channel: 10% crustaceans, 4% small fish, and 7% nemertean worms
- Reservation Point: 13% polychaetes, 27% crustaceans, 4% small fish, and 7% nemertean worms
- Hyperion: 7% crustaceans, 25% small fish, and 7% nemertean worms

It was not possible to obtain sufficient bile from fish sampled at this site in December 1984; accordingly, values obtained from a subsequent sampling (August 1986) are given. Individual compounds in the bile were determined in our laboratories by gas chromatographic/mass spectroscopic analysis. The weight percentages from the analysis of bile from one white croaker from Reserve Point (sampled on August 26, 1986) were as follows:

- Crotex (27%) were also found.

The mean concentrations of metabolites of aromatic hydrocarbons present in the kerosene/gasoline fraction of bile were generally higher than those obtained from the White Point and Hyperion outfall fish. The pattern of values obtained from the Hyperion site was similar to that for BaP; the mean concentrations of metabolites from the Hyperion site were higher than those obtained from English sole from nonurban reference areas in Puget Sound (6, 7, 9, 10); however, NPH values in white croaker were 5-7 times higher.

Liver Lesions in White Croaker. The white croaker subjected to chemical analysis were also examined histopathologically. Of the liver lesions detected, only the most apparently serious conditions are reported here. A more detailed description of the histopathologic characteristics of these lesions will be presented elsewhere (26). The liver lesions included neoplasms, putative preneoplastic lesions (i.e., basophilic focus of hepatocellular alteration), megalocytic hepatosis, and hepatocellular clear pleomorphism (5-7, 18, 22, 23, 26, 27). The types of fish liver neoplasms and the sites from which they were captured are as follows: a cholangiocellular carcinoma in one fish (25 fish examined) from Queensway Bay; a hepatocellular carcinoma in one fish (25 fish examined) from the Los Angeles area. In San Pedro Bay, the summed concentrations of PCBs in livers from the Los Angeles area contained substantially higher concentrations of metabolites of aromatics compounds in fish from the Los Angeles area (except those from the Hyperion site) compared to fish from Dana Point, regardless of whether the values were obtained at BaP or naphthalene (NPH) wavelengths. The values obtained at BaP and NPH wavelengths primarily represent metabolites of polynuclear AHS, characteristic of fossil fuel combustion.

In other fish (20, 30) and mammalian species (31), the probability of developing detectable liver neoplasms, as well as certain other liver lesions, increases with age. Because the presence of white croaker with liver neoplasms at the Los Angeles sites could partially be due to the capture and sampling of older fish rather than the results of exposure to environmental carcinogens, it is important that the age composition of white croakers from reference areas be comparable to that of croaker from urban areas.

In addition, the analyses of bile (Table II) revealed large differences in concentrations of metabolites of aromatic compounds in fish from the Los Angeles area (except those from the Hyperion site) compared to fish from Dana Point, regardless of whether the values were obtained at BaP or naphthalene (NPH) wavelengths. The values obtained at BaP and NPH wavelengths primarily represent metabolites of polynuclear AHS, characteristic of fossil fuel combustion.
Cholangiocellular carcinoma

Differences, for example, in the concentrations of bile
metabolites measured at BaP wavelengths for fish from
Cerritos Channel and Reservation Point were, as indicated,
many times greater than those for Dana Point. These
findings provide clear evidence of the high degree to which
the white croaker form the Los Angeles area had been
exposed to toxic chemicals. Interestingly, the concentra-
tions of aromatic compounds in the bile of fish from
Cerritos Channel and Reservation Point were higher
than those obtained with English sole from Eagle Harbor in
Puget Sound where the sediments were heavily impreg-
nated with creosote AHs (7). In Eagle Harbor, the English
sole had bile values, determined at BaP wavelengths, of
21 ± 1500 ng/g (7).

The observed high degree of uptake of aromatic com-
pounds, such as fossil fuel hydrocarbons, by white croaker
from the Los Angeles area and the contamination of food
organisms with AHs and CHs have not been expected

Chlorinated hydrocarbons have previously been iden-
tified in white croaker from the Los Angeles area. Brown
et al. (4) reported concentrations of DDT and related
compounds similar to our values for livers of white croaker
from White Point and Dana Point (178000 and 2800 ng/g,
respectively, converted to dry weight by using a multiplier
of 5; n = 10 per site). Gossett et al. (12) reported the
concentrations of DDT and related compounds in muscle
of white croaker from White Point and Dana Point to be
38000 and 700 ng/g, respectively. Although studies re-
ported by Young et al. (33) and Bascum (34) demonstrated
that body burdens of DDT's and PCBs increased with
the Los Angeles area and the contamination of food
organisms consumed by white croaker, or the route(s) of uptake of chemicals by
this species, was provided.

The liver neoplasms, preneoplasms, and other liver le-
sions (e.g., megalocytic hepatosis) in white croaker, dem-
strated here for the first time, resemble those found in
bottom-feeding fish from other polluted coastal areas (5-7,
19, 20, 27). Furthermore, the megalocytic hepatosis, as well
as the neoplastic and preneoplastic liver lesions in the
white croaker, compares morphologically to those identi-
fied in laboratory animals exposed to toxic and/or carci-
nogenic chemicals (21, 28). Overall, the chemical and
biological findings suggest a possible relationship between
toxic environmental chemicals and the observed liver le-
sions; however, on the basis of previous data from Puget
Sound (2, 5-7, 10), it was surprising to find relatively low
prevalences of neoplastic and preneoplastic liver lesions
in the white croaker from the Los Angeles area. Clearly,
more work is needed to determine if these differences are
species-related and/or attributable to other factors. In
this regard, we do not feel that seasonal variability was a
significant factor in the differences observed. For example,
there is no reason to believe on the basis of present evi-
dence that tissue concentrations of xenobiotics would vary
significantly over a 3-month period. Moreover, for the five
sites sampled in a single month (December), substantial
differences were found in concentrations of chemical
contaminants in sediments and fish, as well as in preva-
lences of fish diseases.

The benthic invertebrates eaten by the Los Angeles fish
had apparently bioconcentrated AHs and CHs present in
polluted sediments. That is, the concentrations of chem-
icals in the food organisms were generally higher than those
in the sediment. For example, food organisms of the fish
from Cerritos Channel had concentrations of summed AHs,
PCB, and DDT-related compounds that were 34, 8 and 58
times those in the sediment, respectively.

The observed contamination of the fish through the
consumption of benthic food organisms revealed an im-
portant route of exposure to toxic chemicals; however,
other routes are possible—both AHs and CHs are known
to be bioconcentrated by fish from the sediment (35, 36)
and water column (16, 37, 38). Undoubtedly, the relative
impact on the croaker of the various routes of exposure
will have to await further studies in the field and labora-
tory.

Clearly, the complementary use of chemical analytical
data on stomach contents, bile, and liver of the white
croaker, in conjunction with histopathologic examination
of the liver, has provided a fundamental case for the study
of
Moreover, in the broad sense, the findings reported here heighten interest in the extent of contamination of food chain organisms in urban coastal environments and the consequences to higher forms of marine life and to humans.

Acknowledgments


Registry No. DDT, 56-29-3; o,p'-DDD, 3424-82-6; p,p'-DDD, 72-56-9; phenanthrene, 85-01-4; fluoranthene, 206-44-0; pyrene, 129-00-0; phenanthrene, 85-01-3; 1-methylphenanthrene, 832-69-9; benzo[a]pyrene, 50-32-8; perylene, 198-55-0; dibenz[a,h]anthracene, 52307-64-9; chrysene, 218-01-9; benzo[e]pyrene, 72601-64-9; heptachlorobiphenyl, 28655-71-2; octachlorobiphenyl, 26914-33-0; heptachlorobiphenyl, 28655-71-2; octachlorobiphenyl, 55722-24-4; nonachlorobiphenyl, 53742-07-7.

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