

**2,3,7,8-Tetrachlorodibenzo-p-Dioxin (TCDD) and 2,3,7,8-Tetrachlorodibenzo-p-Furan (TCDF)
In Blue Crabs and American Lobsters from the New York Bight**

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ABSTRACT

An investigation of 2,3,7,8-tetrachlorodibenzo-p-dioxin (TCDD) and 2,3,7,8-tetrachloro dibenzo-p-furan (TCDF) bioaccumulation in blue crabs and American lobsters collected from Newark Bay and the New York Bight revealed widespread contamination across the continental shelf. Separate analyses of muscle and hepatopancreas tissue identified the presence of TCDD and TCDF in the latter tissue only. Analyses of the edible tissue (combined muscle and hepatopancreas) displayed varying concentrations across the ranges of both organisms. Blue crabs from the winter dredge fishery area, at their seaward migratory limit, displayed the highest levels (TCDD = 73 pg./g; TCDF = 67 pg./g). American lobsters sampled beyond the ocean dumpsites for harbor dredge spoils have slightly higher levels of TCDD (Mean: 40 pg./g) than inshore, Ambrose fishery lobsters (31 pg./g) and offshore lobsters caught near the edge of the continental shelf. Some locational differences are caused by offshore dumping of dredge spoils and are shown to fluctuate seasonally due to the migratory movements of the lobsters. The additive toxicity of TCDF was calculated as TCDD equivalents, summed with the measured TCDD and compared to FDA "Levels of Concern" and revealed unacceptable levels of risk associated with the consumption of these organisms.

INTRODUCTION

In 1984, dioxin contamination of soils was discovered at a pesticide-manufacturing site on the Passaic River in New Jersey prompting a study of sediments and biota both upstream to the head of tide and downstream into Newark Bay and the New York Bight (1). Sixty percent of the sediments sampled from the river, adjacent to the facility, showed detectable levels of 2,3,7,8-tetrachloro dibenzo-p-dioxin (TCDD) with higher levels being found upstream and at depth possibly due to the flood dominated, salt-wedge dynamics in the estuary. It is estimated that substantial quantities of this contaminant are in storage within the sediments of the river and its downstream embayment as well as at the dredge spoil dumpsite, 12 miles offshore, in the New York Bight (Figure 1).

A wide range of finfish and crustacea from the river showed tissue levels of TCDD (Figure 1a). Blue crabs from the lower estuary contained the highest concentrations (Mean: 468 pg./g in hepatopancreas and 22 pg./g in muscle). Several migratory species, blue crabs and striped bass from the downstream embayments and American lobsters from nearshore ocean waters also showed elevated levels of TCDD. American lobsters from the New York Bight showed consistently high levels of TCDD in the edible hepatopancreas (Mean: 77 pg./g) and combined muscle and hepatopancreas (Mean: 44 pg./g). These lobsters were collected at the mouth of New York harbor and approximately twenty miles offshore beyond the ocean waste disposal sites for dredge spoils, fly ash and sewage sludge.

The identification of TCDD in blue crabs and lobsters distant from the only apparent source suggested a more widespread zone of contamination and a potentially higher human health risk due to the intensive amount of commercial fishing in these offshore waters. The objectives of the present study were to: 1.) Investigate other potential areas of TCDD impact by sampling the seaward migratory limit of the blue crab (i.e., Raritan Bay) and the entire migratory range of the American lobster within the New York Bight (i.e., up to 150 miles offshore); 2.) Estimate the relative effects of location and season on lobster contamination due to their migratory behavior; 3.) Identify the presence of TCDF in the animals (which shows an equivalent level of toxicity compared to TCDD); 4.) Convert any levels of TCDF found into TCDD equivalents for summation of the total risk as compared to the Food and Drug Administration's (FDA) "Levels of Concern" for TCDD; and to 5.) Investigate the relative role of the Historical Area

Remediation Site (HARS) or ocean dredge material dumpsite as a possible intermediary in the transport of TCDD and TCDF to ocean waters and biota.

EXPERIMENTAL SECTION

Sampling

Blue crabs were collected by the use of crab pots, otter trawls and purchases from commercial Crabbers. Sites sampled included locations from Raritan Bay, Sandy Hook Bay and upper New York harbor. American lobsters were collected in lobster pots and otter trawls for Raritan and Sandy Hook Bays or purchased from commercial lobstermen for deepwater areas. Lobster catch locations were sub-divided into three fisheries as defined by NMFS, the National Marine Fishery Service (2), which generally reflect different ports of departure for the fishing fleets operating in these waters (Figure 2). The Ambrose fishery includes Raritan and Sandy Hook Bays extending to a hypothetical 7 nautical mile radius on the Ambrose Light leading into New York Harbor. This area also includes the blue crab sampling locations previously mentioned. The Alongshore fishery is a box-like area extending from Long Branch to Point Pleasant and extends offshore approximately 25 nautical miles following the Hudson Canyon. This area includes the ocean waste disposal sites for dredge spoils, fly ash and sewage sludge (Note: this sewage sludge dump is now closed and material is now deposited 106 miles offshore). The Offshore fishery extends eastward from the 50-fathom line to the 100-fathom line approximately 100 miles seaward to the edge of the continental shelf.

Analysis

Samples for analysis consisted of standardized edible portions (Figure 2a) including the thoracic, claw, leg and tail meat as well as the hepatopancreas combined (i.e., worst case exposure). Tissue from five organisms of similar size and weight were composited and homogenized in a food processor to generate comparable amounts of material which was then held frozen until extraction. In order to determine if differential bioaccumulation was occurring, a small number of analyses were performed on single lobsters in which the muscle and hepatopancreas tissue were processed separately (Figure 2b).

Tissue analyses were performed using a modified EPA method (3) with high-resolution gas chromatography, low-resolution mass spectroscopy. The modification included a saponification of the tissue prior to the initial extraction. Clean up involved passing the extract through a series of five columns with the final step involving an activated carbopak/elite mix in a 2-cm column with final TCDD and TCDF elution by toluene. Before clean up the samples were fortified with C₁₃ labeled TCDD and TCDF as an internal standard. The extracts were analyzed using an electron impact GC/MS instrument with a direct capillary interface, and a 60-meter isomer specific fused silica capillary column. If TCDD or TCDF were not detected, a detection limit was calculated based on a 2.5 times signal-to-noise ratio at the retention time of the respective contaminant and the C₁₃ labeled internal standard.

The QA/QC procedures followed EPA guidelines (4) and included spiking tissue of each species with appropriate standards, analyzing replicate and blind control samples and demonstrating the proper isomer specificity and ion ratios. The mean percent recovery for spiked samples with internal standards was 96.8 % with +/- 1 % error for the full range of analyses. The actual values reported in the tables are not corrected for recovery.

Statistics

All data are reported as arithmetic means with the method detection limit of 10 pg./g used for all non-detectable values in summation and statistical analyses. Since it is not possible to catch lobsters at all locations and seasons due to life history characteristics and sampling limitations, it proved difficult to test the significance of any space/time interactions on a strictly monthly or seasonal basis. Knowing that the animals do perform a regular onshore/offshore migration, however, it was possible to separate the data into Spring-Summer (Onshore) and Fall-Winter (Offshore) migration periods. These spatio/temporal differences were tested for significance via a 2-Way ANOVA (5) for unbalanced design on raw and ranked (i.e., non-parametric) data. The model used Season and Location as classes and TCDD and TCDF as the dependent variables. A significance level of 0.05 was used for all hypothesis testing concerning the F statistic.

The data for TCDF was converted to "TCDD Equivalents" via the EPA Toxic Equivalency Factor Method (6) recommended to quantify the additive risk from this contaminant. The results were then compared to the FDA recommended "Levels of Concern" for TCDD in order to estimate the level of unacceptable risk (7). These recommendations are: 1.) No consumption for levels greater than 50 pg./g; and 2.) Reduced consumption for levels between 25 and 50 pg./g.

RESULTS

Blue Crabs

Blue Crabs tissue (i.e., muscle and hepatopancreas combined) from Raritan Bay and the lower Hudson River (Table 1) show mean TCDD concentrations of 73 pg./g (Range 10-260 pg./g) and TCDF at 67 pg./g (Range: 10-110 pg./g). When TCDF is converted to TCDD equivalents the actual concentration increases to 80 pg./g. A large percentage of the captured animals tested positive for both TCDD (53%) and 2,3,7,8-TCDF (73%) with the latter being more ubiquitous. An analysis of the distribution of TCDD and TCDF in Blue Crabs across the harbor (Figure 3) shows that the contaminants are usually found together and an apparent increase in concentration occurs from inner Raritan Bay out to the mouth of the Hudson River. All of the control samples from Delaware Bay showed no detectable levels for either contaminant. The TCDD levels for these Blue Crabs collected in 1986 (at the seaward limit of their migratory range) were similar to levels found (1) within Newark Bay in 1983 (Figure 3a).

American Lobsters

Separate analyses of lobster muscle and hepatopancreas showed that both of the contaminants were present only in the hepatopancreas (Range: TCDD= <10-290 pg./g; TCDF= <10-320 pg./g). However, they may be present at levels below the relatively high detection limits in this study. Analyses of combined muscle and hepatopancreas for the entire New York Bight lobster fishery (Table 1) show mean TCDD concentrations of 28 pg./g (Range: <10-110 pg./g) and TCDF at 26 pg./g (Range: <10-120 pg./g). When TCDF is converted to TCDD equivalents the actual dosage increases to 30 pg./g. In contrast to the crab data, a smaller percentage of the lobsters tested positive for both TCDD (36%) and TCDF (20%) with the former being more ubiquitous. This may reflect the more remote exposure to the source of contamination (Passaic River) or may be due to the large number of lobster analyses showing high detection limits (i.e., >25 pg./g).

If the New York Bight lobster results are broken into sub-fisheries extending progressively offshore as categorized by NMFS (2) the data reveal some significant locational differences (Figure 4). TCDD and TCDF are high in both the Ambrose and Alongshore fisheries but low to non-detectable in the far offshore areas (Figure 4a). The 15 pg./g TCDD for Offshore lobsters is based on only 2 positive results out of 17

composites. The results illustrate that levels of TCDF appear to drop off linearly as one progresses offshore. However, this differs dramatically from the TCDD levels, which are higher in the Alongshore area, near the dredge spoil dumpsite, than in either, the Ambrose or Offshore fishery areas. These spatial differences may be related to seasonal feeding patterns and migratory movements.

In Figure 5a the monthly lobster landings for New Jersey waters (N.J. Div. Fish, Game and Wildlife) are superimposed on the mean monthly TCDD results from the 1985-1986 sampling season. The annual commercial landings appear unimodal. They begin to increase in early spring (March) as water temperatures increase and the lobsters become more active and begin their shoreward migration. The commercial fishing activities also increase at this time, as the lobsters are actively feeding and the milder weather conditions are more suitable to commercial lobstering. The landings reach a peak in late summer (August) and steadily decline thereafter as the water temperature drop and the animals seek deeper water to avoid severe winter conditions.

In contrast the TCDD data for the New York Bight fishery appears bimodal with high values in the winter/early spring followed by a steady rise in contamination from May through September. The latter trend probably reflects the lobster's active feeding and growing phase but does not explain the high levels found in the winter and early spring. These anomalies results become less contradictory if the data are separated into Ambrose and Alongshore areas and examined with an understanding of the seasonal, migrational movements of the lobsters within these sub-fisheries.

The Ambrose lobsters (Figure 5b) consistently had non-detectable levels of TCDD through the spring and summer but then rose remarkably high in the fall and winter. The Alongshore lobsters on the other hand (Figure 5c) had elevated levels of dioxin in all four seasons with an apparent increase in late fall. These trends were similar for both the 1985 and 1986 samplings seasons.

Spatial partitioning for TCDD (Figure 4) originally indicated that levels were higher in the Alongshore than in the Ambrose fishery. Migrational analysis (Figure 6a) reveals that this relationship only holds for the Spring-Summer (onshore migration) period and that by the Fall-Winter (offshore migration) this trend is reversed with higher levels inshore and progressively lower values as sampling moved offshore. Spatial partitioning for TCDF (Figure 4) originally indicated that the highest concentrations were found inshore and that progressively lower levels occurred offshore. Migrational results (Figure 6b) reveal that this relationship only holds for the Fall-Winter (offshore) period and that by the Spring-Summer (onshore migration) the data resemble the TCDD results with higher levels of contamination Alongshore than in the Ambrose fishery. Therefore, on a seasonal/migratory basis the levels and distribution of these two contaminants seem to fluctuate in a similar pattern.

This relationship is borne out statistically by the ANOVA which shows significant differences between location and seasonal effects on the concentrations of both TCDD ($P > F = 0.001^*$) and TCDF ($P > F = 0.03^*$) in American lobsters. Significance results were identical for both raw and ranked data so only the raw results are reported here. However, when the variances are partitioned into a partial sum of squares, season becomes non-significant for TCDD while location ($P > F = 0.02^*$) and the interaction term ($P > F = 0.003^*$) remain significant. The levels of TCDF on the other hand show non-significant effects in the partial sum of squares for location, season and the interaction term.

DISCUSSION

The Blue Crab has only a three year life span which begins as a microscopic larva that is spawned into the nearshore ocean waters. They then undergo a series of sequential metamorphoses, molts and movements back into the estuary where they exist as adults until the females once again migrate seaward to spawn (8). Therefore the animals that were found contaminated in Raritan Bay in 1986 had just entered the estuary from their offshore planktonic stage when the original 1983 investigation was initiated. This indicates that the contamination is not only transported out of the estuary into the nearshore food chain but that it is persistent over time. In addition the amount of TCDD found in these crabs exceeds the FDA's "Level of Concern" recommending no consumption (i.e., >50 pg./g). The presence of TCDF also adds to the risk as indicated by its conversion to TCDD equivalents.

American lobsters in the New York Bight exhibit high yet varying levels of both TCDD and TCDF across their entire geographic range. Although low levels of TCDF congeners have been reported for American lobster hepatopancreas in the past (9) this was the first evidence of TCDD and high TCDF contamination. In addition, the broad distribution of contamination across the New York Bight appears unique compared to other reported contamination events for this species which tend to be localized in the nearshore (10) or estuarine areas (11).

The statistical analysis supports the presence of spatio/temporal differences in TCDD and TCDF contamination in American Lobsters across the New York Bight section of the continental shelf. The lack of significance for TCDF in the partial sum of squares analysis may be due to the unbalanced design and large variances inherent in the data. For TCDD the partitioning analysis indicates that location is more important than season although the significant interaction term suggests that the spatial effects are strongly dependent on the time of year in some unspecified way. Biologically the interaction term may be synonymous with the definition of migratory behavior, which manifests itself as changes in spatial relationships based on temporal cues during the annual life cycle of an organism.

The significance of the location effect and the interaction term may be better understood if we ignore the three artificial fishery boundaries and consider the lobsters as biological populations. Field studies indicate that regardless of latitude the seaward American lobster populations for the eastern continental shelf consist of two sub-populations (12); one that remains in the nearshore area and moves at most 15 km across a home territory and a second deepwater population that resides much further offshore and performs true long-range migration. The seasonal migration is possibly associated with maximizing degree-days for molting, growth, gonadal development, and egg extrusion (13). In addition, the mature lobsters on average move significantly greater distances than immature lobsters with the latter tending to travel along the coastline whereas the larger more mature animals move seaward and to greater depths, as they grow older.

Therefore, the Ambrose and Alongshore locations may circumscribe two ends of the same inshore lobster territory with a size-stratified population moving to variable directions and depths and being exposed alternately to two sources of contamination - the Hudson River plume and the HARS (i.e., ocean dredge spoil disposal site). Moving slowly inshore during the spring/summer period the deepwater, more mature lobsters will mix with the smaller inshore population. Animals in the entire Ambrose fishery then bio-accumulate higher levels of TCDD and TCDF during this more active phase until the mature animals move offshore in the Fall. Then they either depurate some levels during over-wintering at the Alongshore area and/or mix with the landward edge of the less contaminated offshore population resulting in lower

average levels over the winter. In contrast smaller lobsters within the Ambrose fishery aestivate during the winter by burying themselves in the soft muddy channels which criss-cross the Lower Bay. During this time they metabolize much of their stored fat and may release any associated contaminants.

Although it is difficult to separate out the proximate causes of the observed spatio/temporal variations in TCDD and TCDF contamination, we can assume that it includes differences in source exposure, activity cycles, lipid metabolism and possibly depuration. The seasonally persistent high levels of contamination at the seaward end of the inshore population's range (i.e., the Alongshore area) strongly suggests that dredge spoils were supplying a continuous source of TCDD and TCDF to American lobsters. In support of this observation is data from a subsequent study of Newark Bay blue crabs, Alongshore lobsters and soils from the manufacturing site on the Passaic River (14) which reveals similar GC/MS fingerprints of other dioxin and furan congeners between animals and sites.

The amount of TCDD in the New York Bight fishery and more specifically the inshore Ambrose and Alongshore fisheries exceeds the FDA's "Level of Concern" recommending reduced consumption (i.e., >25 pg./g). If TCDF is converted to TCDD equivalents the actual dosage increases in the Alongshore fishery towards a recommendation of no consumption. The large number of lobster analyses showing high detection limits (i.e., 28 % >25 pg./g) may also mask more contamination. This is especially critical when an advisory level exists in the low parts per trillion range. Unfortunately this is a common analytical problem for tissue samples requiring picogram per gram sensitivity since the presence of other organochloride contaminants such as PCBs may mask the presence of both TCDD and TCDF (15). PCBs are a common contaminant in lobsters along the entire eastern seaboard including the New York Bight (1) Long Island Sound (16) and New Bedford Harbor (11). In addition, the conversion to TCDD Equivalents of the other PCDD and PCDF congeners recently found to bioaccumulate in these animals (14) reveals a much higher level of risk than expected from the TCDD and TCDF analysis presented here, although most of the risk comes from the highly toxic TCDD and TCDF.

Concerning the safety of New York Bight lobsters as a food source it is apparent that a threat does exist but the presence of the contaminants in a secondary part of the edible tissue (hepatopancreas) gives health officials some latitude in advising the public concerning the risks. The risk to consumers could be much reduced by not eating the hepatopancreas. In addition, separating the hepatopancreas before cooking (i.e., cooking lyses the organ and disseminates the contamination (10)) can significantly reduce the potential exposure to this contaminant. In addition a recent study has shown that TCDD residues in fish may be reduced through selective cooking and processing techniques (17). A similar study is needed to address the way that lobsters and crabs are usually cooked (i.e., boiled whole). It is to our benefit, considering the Earth's dwindling natural resources, and its ever-increasing human population, to preserve and protect these important food species.

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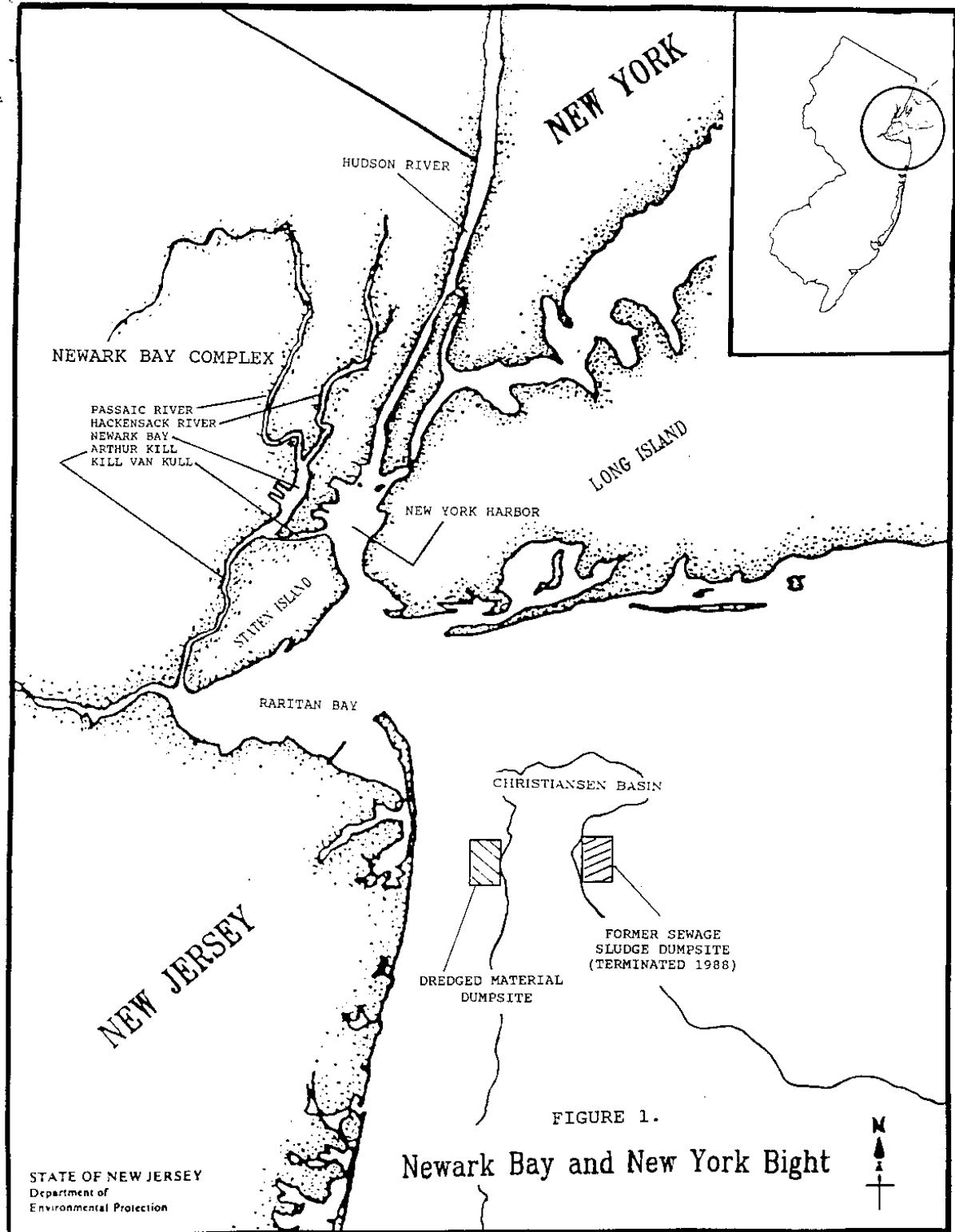
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Table 1. TCDD, TCDF and TCDD Equivalent¹ Levels in Blue Crabs and American Lobster From Raritan Bay and the New York Bight

Blue Crab	X	TCDD ² S.D.	n	%Pos	%> 25pg/g ⁴	X	TCDF S.D.	n	% Pos	TCDD Equivalents X
	73.1	(82.9)	15	53	60	70	(36.4)	15	73	80
American Lobster³										
New York Bight	27.5	(25.7)	47	36	36	26	(28.9)	39	20	30
Ambrose	31.2	(30.2)	19	37	37	35.0	(35.0)	19	37	34.7
Alongshore	40.5	(22.8)	11	73	73	24.5	(19.5)	11	36	42.9
Offshore	15.0	(13.9)	17	12	12	<10	-	9	0	15.0

1. After Barnes et al 1986
 2. Arithmetic means include all non-detectable values at 10 pg/g
 method detection limit for summation SD = standard deviation N = no
 of 5 organism composites analyzed % Pos = Percent Positive
 3. Lobsters data presented for NY Bight as a whole and its sub-
 fisheries 4. FDA "Level of Concern" recommending reduced consumption
 is 25 pg/g



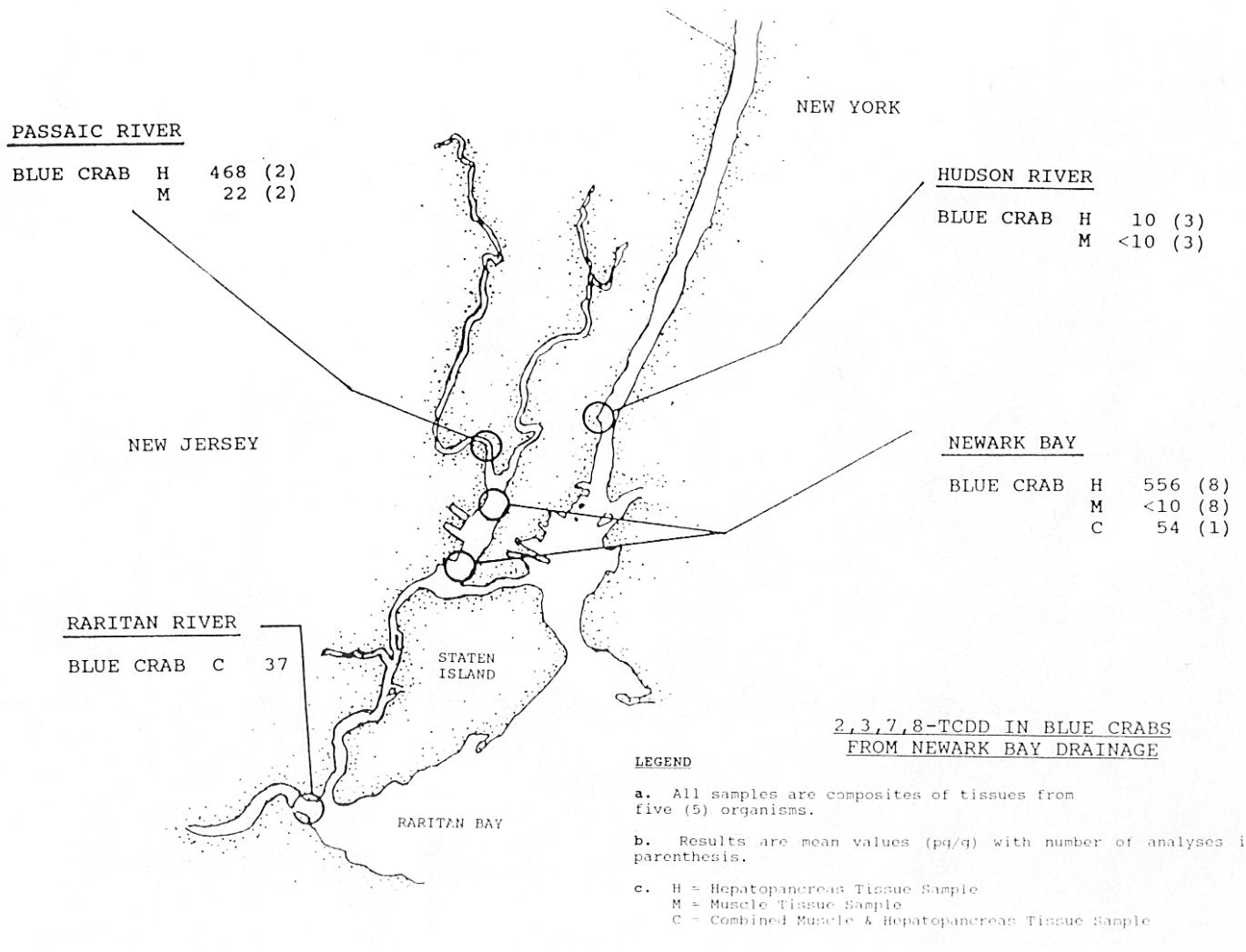
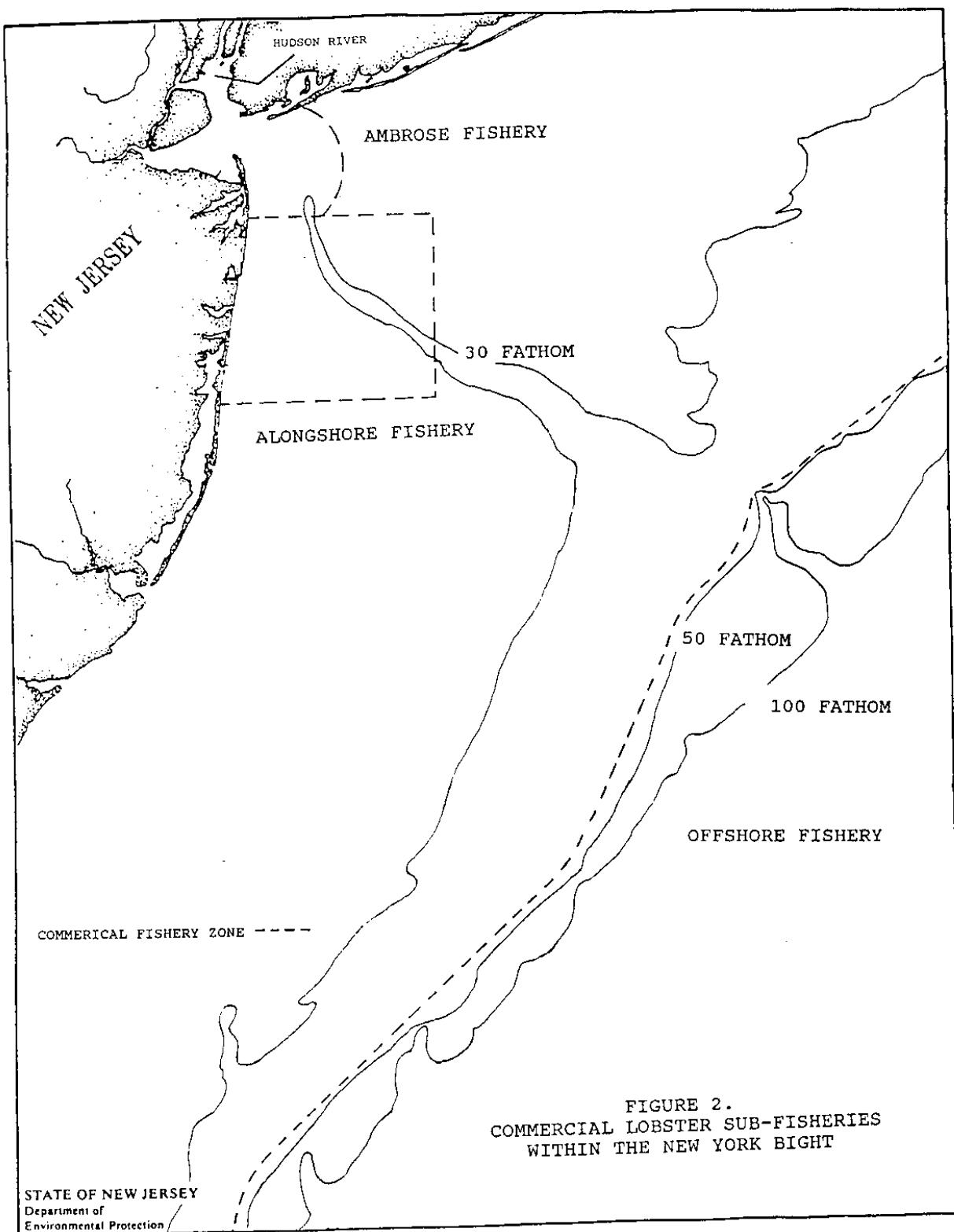


Figure 1a. Dioxin (2,3,7,8,-TCDD) in Blue Crabs From Newark Bay 1984*

From Belton et al. 1985



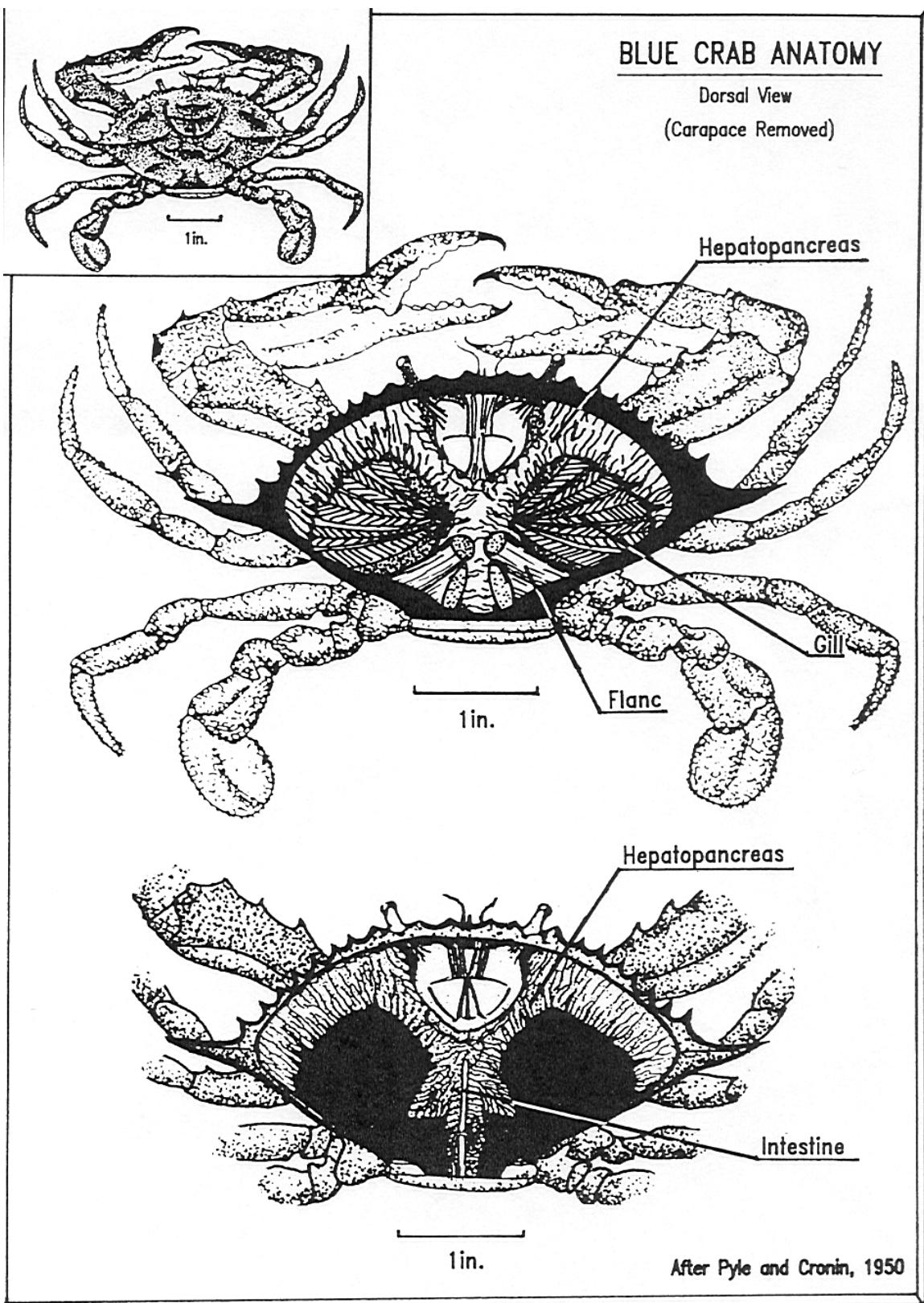


Figure 2a. Blue Crab Anatomy

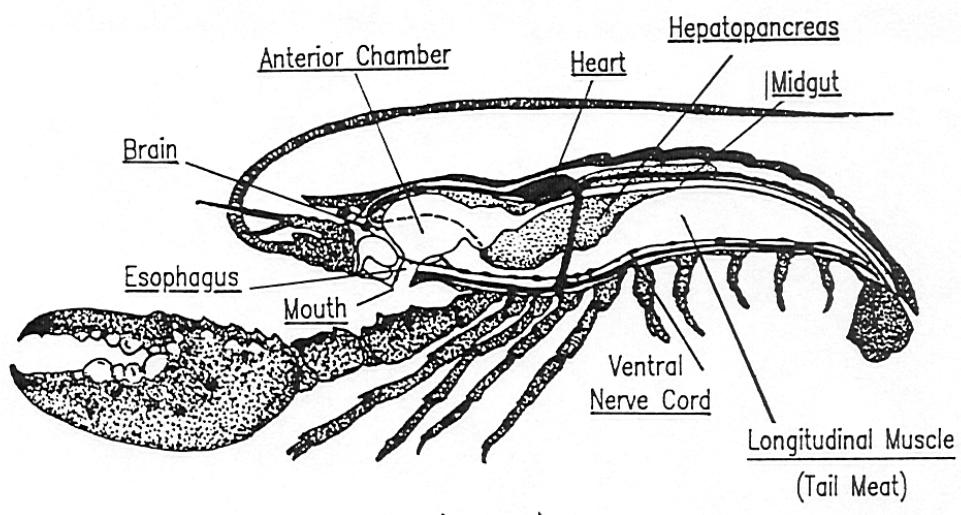
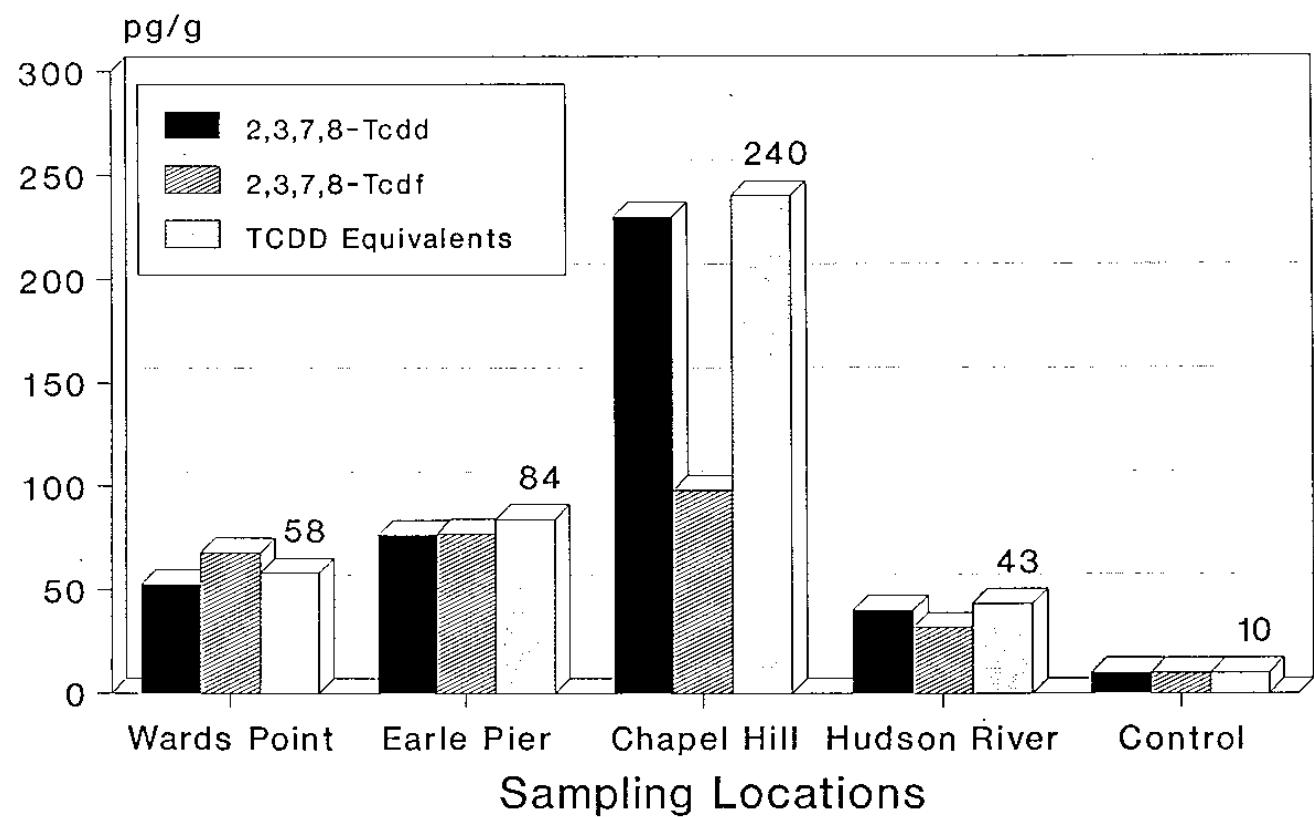


Figure 2b. American Lobster Anatomy

Figure 3. 2,3,7,8-TCDD, 2,3,7,8-TCDF and TCDD Equivalents in Blue Crabs from Raritan Bay and the Lower Hudson River



TCDD Equivalents via Barnes et al 1986
Controls are from Delaware Bay,
10 pg/g is the method detection limit.

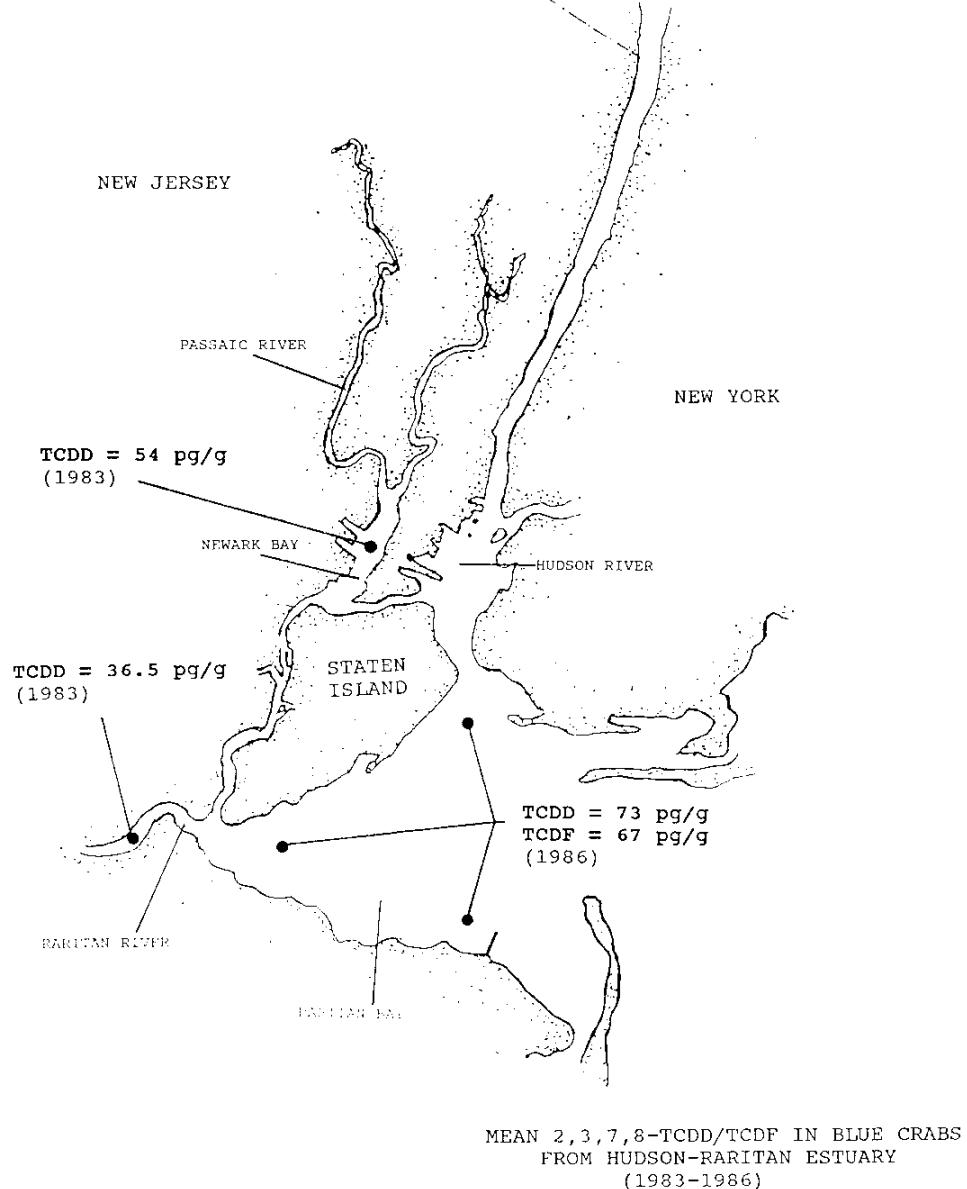
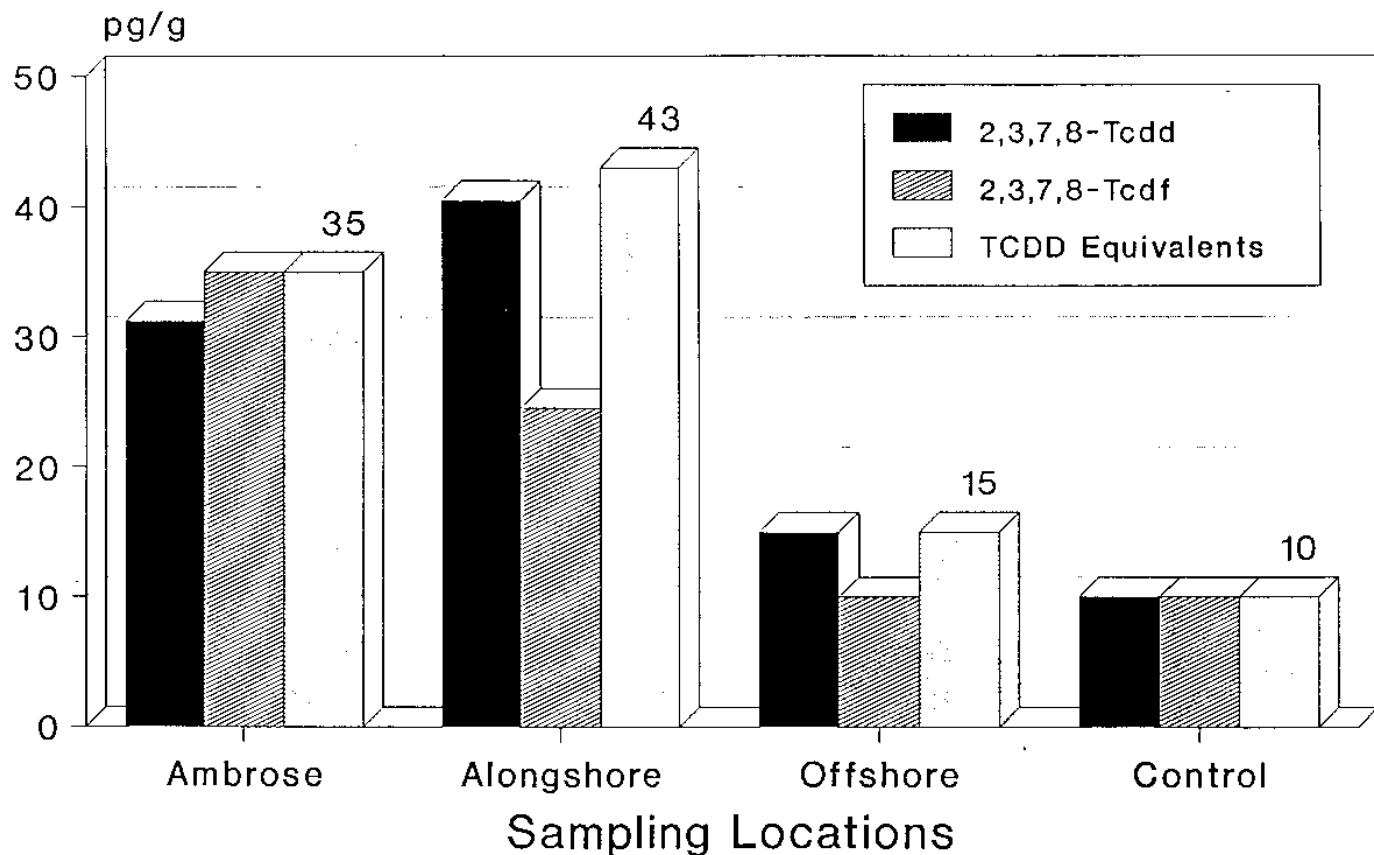


Figure 3a.

Figure 4. 2,3,7,8-TCDD, 2,3,7,8-TCDF and TCDD Equivalents in Lobsters from Sub-Fisheries in the New York Bight



TCDD Equivalents via Barnes et al 1986.

Controls are from Maine.

10 pg/g is the method detection limit.

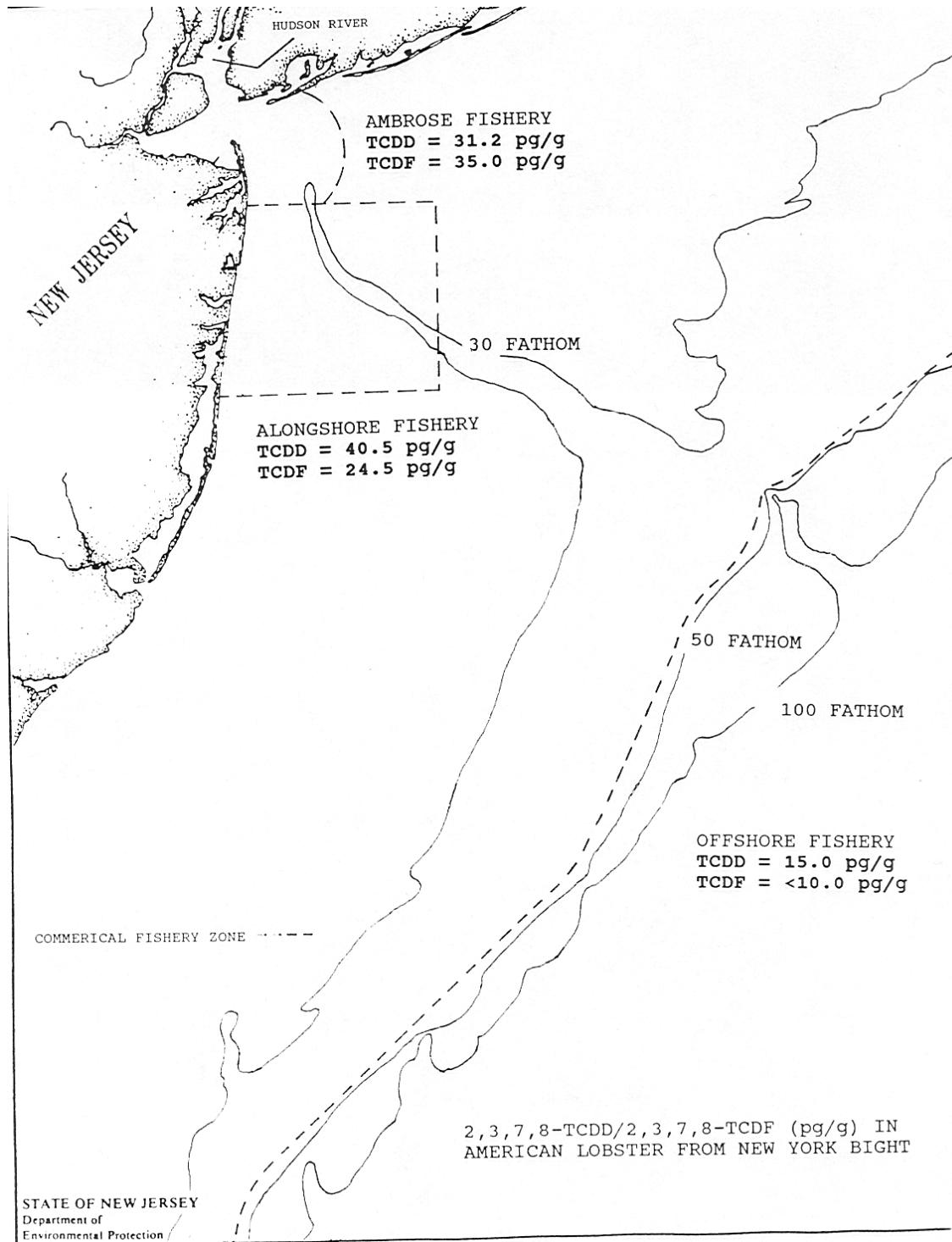


Figure 4a.

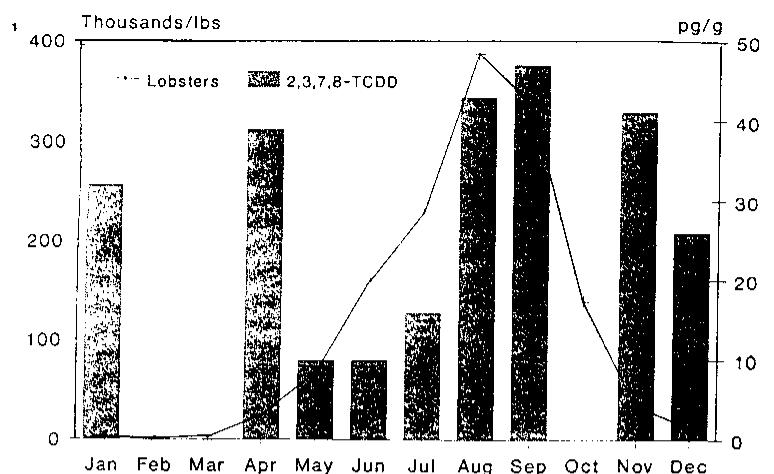


Figure 5a. Landings and 2,3,7,8-TCDD Levels in New York Bight Lobsters

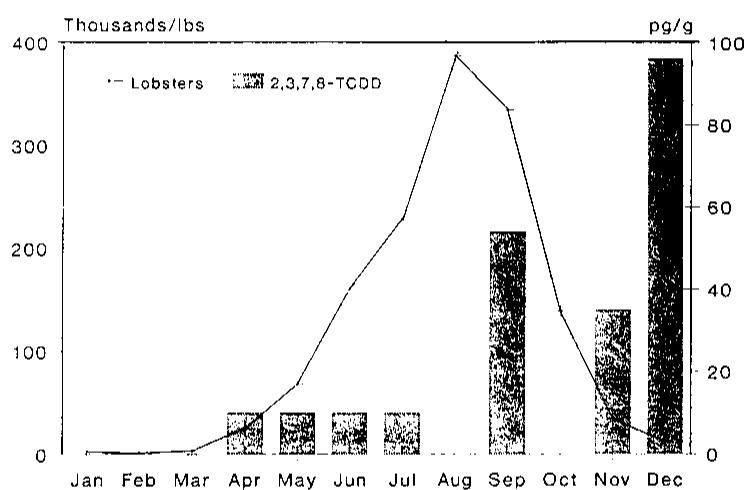


Figure 5b. Ambrose Fishery

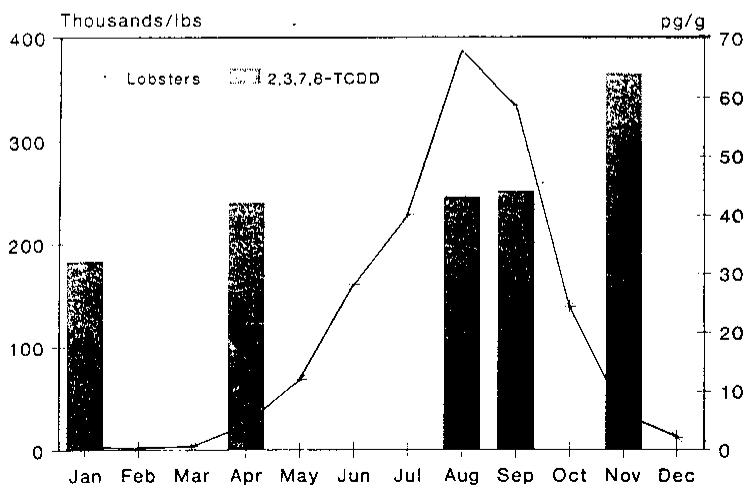


Figure 5c. Alongshore Fishery

FIGURE 5a,b,c.

Figure 6a. Mean 2,3,7,8-TCDD in Lobsters
by Location and Migration Period

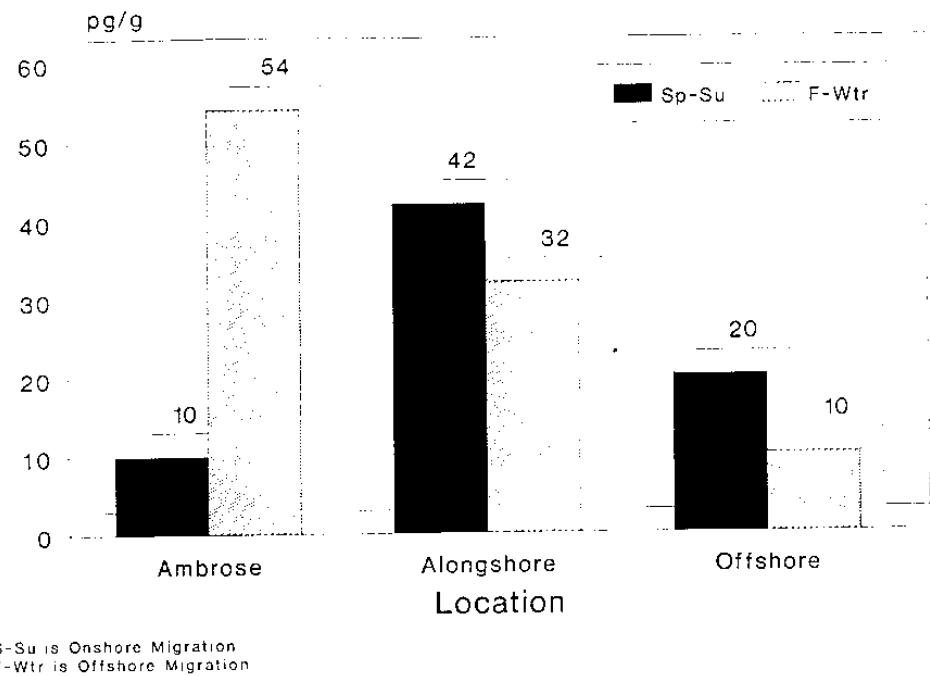


Figure 6b. Mean 2,3,7,8-TCDF in Lobsters
by Location and Migration Period

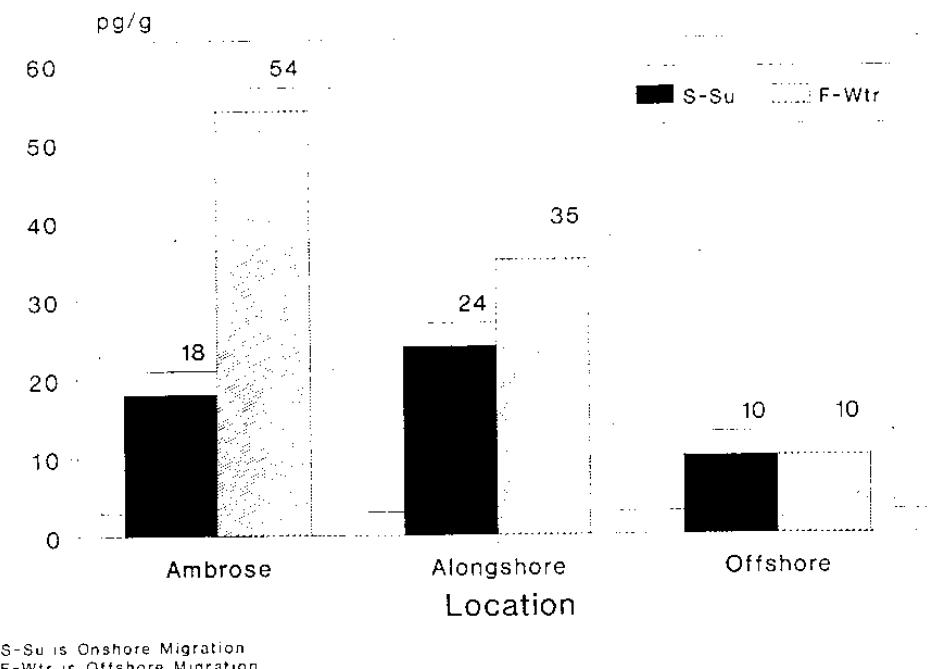


FIGURE 6a,b