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Environmental Contamination and Toxicology

Contaminants in Recreationally Important Estuarine Finfish from South Carolina

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The South Carolina coast has been both rapidly and widely developed in the last two decades. As the coastal population has increased, the potential for pollution has increased correspondingly. Although point-source pollution from industry and sewage treatment plants is regulated, nonpoint sources for the most part are not and are probably increasing as development expands along the coast.

In general, much work in the Southeast has been directed towards contaminants in water, filter feeders, and sediments (Marcus and Mathews 1987 and Shuster and Pringle 1969). The reasons for such studies are quite valid, since organisms usually are immersed in water constantly, and both sediments and filter feeders tend to concentrate contaminants. Examining a broader range of organisms and species is necessary, however, to determine possible bioconcentration throughout the estuarine biota. Although contaminant work utilizing various finfish has been conducted in many areas, relatively few such studies have been done in South Carolina estuaries, especially with the recreationally-important species of this study (Stickney et al. 1975 and Winger et al. 1990).

As a consequence of the above, this study was initiated by the South Carolina Wildlife and Marine Resources Department (SCWMRD) in 1990. Public support was enlisted via sportfishermen, who assisted SCWMRD in the collection of samples and the identification of possible sources of contamination, e.g. sewage outfalls, industrial effluents, and golf course runoff.

MATERIALS AND METHODS

Red drum (*Sciaenops ocellatus*), flounder (*Paralichthys lethostigma*), and seatrout (*Cynoscion nebulosus*) were collected from 1990-1993 primarily during the warmer months (April through November) when the target species were present in South Carolina estuaries. Samples were collected primarily by ongoing SCWMRD programs (seining and hook-and-line sampling) with help from recreational fisherman (hook-and-line sampling). During the first year whole fish were analyzed, but due to the reluctance of fisherman to submit large fish, small fillets and some livers were utilized the following years. Individual fish samples were ground in a commercial food processor, stored in glass jars (previously cleaned with hydrochloric acid, isopropanol, and distilled water and sealed with teflon-lined caps) and then frozen at -15^0 C until analyzed. Total numbers analyzed were as follows: 78 flounder, 58 seatrout, and 47 red drum for organics and 74 flounder, 42 seatrout, and 66 red drum for

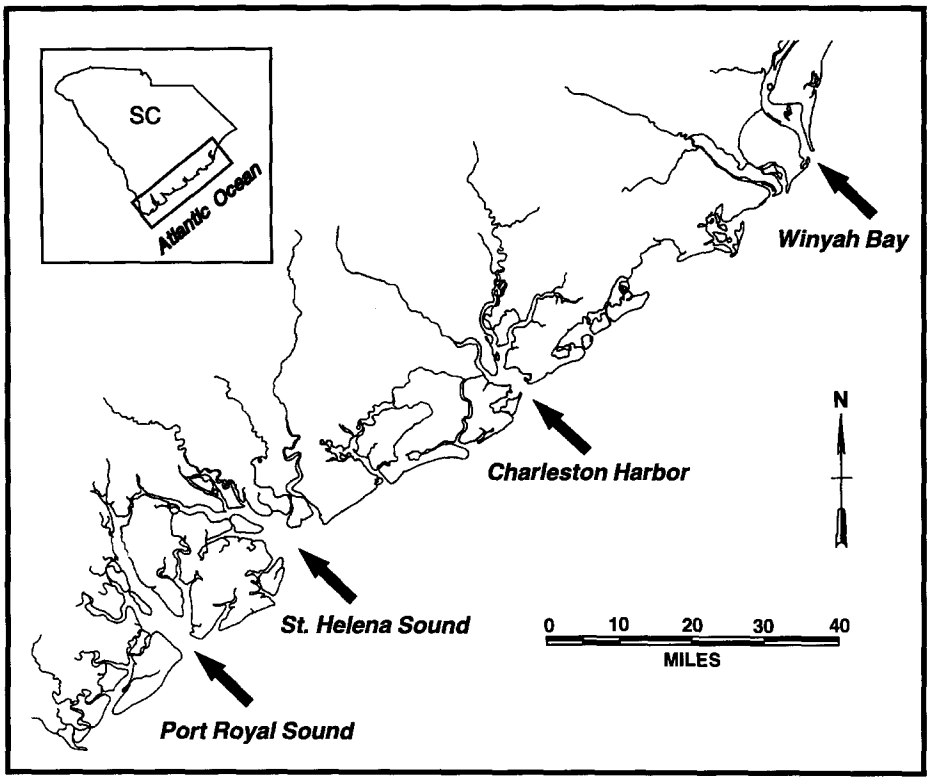

Figure 1. Sampling areas.

metals. (The analysis of each metal on every sample was not always possible due to the small size of many samples.)

Main sampling areas (Figure 1) were selected to cover the major South Carolina estuaries, both highly impacted by development (Charleston Harbor and Winyah Bay) and relatively pristine (St. Helena and Port Royal Sounds). Actual stations within each estuary were determined largely by the availability of specimens. Due to existing programs more samples were obtained from Winyah Bay and Charleston Harbor, whereas the sounds were largely sampled by sportfisherman.

A nitric-acid digestion for the non-volatile metals, copper (Cu), cadmium (Cd), chromium (Cr), lead (Pb), arsenic (As), and nickel (Ni), was performed in a specially-designed microwave oven according to National Institute of Standards and Technology (NIST) methods (Kingston and Jassie 1986). Mercury was prepared by a low temperature digestion, using nitric and sulfuric acids (U S Environmental Protection Agency 1981). An Instrumentation Laboratory Model 751 atomic absorption spectrophotometer (AAS) was used for analyses of all metals except As, which was run by a commercial laboratory.

Metal recoveries were based on spiked samples or NIST bovine liver, one aliquot of which was included with every batch of ten samples and a reagent blank. Percent recoveries were as follows: Cr-69.5 ± 5.2, Cu-83.3 ± 9.7, Pb-78.8 ± 11.3, Ni-74.6 ± 10.2, Cd-85.7 ± 9.2, As-

81.7 ± 9.9, and Hg-88.0 ± 10.6.

Analyses of pesticides (aldrin, dieldrin, endrin, endosulfan, heptachlor, chlordane, and the BHC and DDT series) and PCBs were based on solid phase extraction (SPE) techniques, using 500-mg C18 extraction columns. The samples were extracted with a 2:1:1 solution of methanol, isopropanol, and acetonitrile or acetone. Acetone produced higher recoveries and fewer problems with emulsions, when preparing fatty tissue, and was utilized after the first year. A 15% solution of the extracted sample with distilled water was then poured through pre-rinsed C18 cartridges. The cartridges were rinsed successively with a solution of CH_2Cl_2, hexane, and ethyl ether; methanol; and 15% extraction solvent. After drying, the cartridges were eluted with 1:1 isooctane and ethyl ether. The ether was evaporated prior to bringing the sample up to a constant volume with isooctane. Analyses were by electron-capture gas chromatography on a Tracor Model 540 gas chromatograph, utilizing polar (SGE BP-10) and nonpolar (SGE BP-5) capillary columns for confirmation.

Pesticide recoveries were based on spiked samples, one of which was included with each batch of ten samples and a reagent blank. Percent recoveries were as follows: alpha BHC-86.2 ± 15.9, beta BHC-70.5 ± 7.0, gamma BHC-86.8 ± 10.7, delta BHC-85.4 ± 14.7, heptachlor-80.3 ± 8.8, heptachlor epoxide-83.2 ± 11.1, aldrin-74.7 ± 6.0, endosulfan I-71.9 ± 11.7 and II-68.9 ± 5.7, alpha chlordane-69.9 ± 6.7, gamma chlordane-83.1 ± 8.9, dieldrin-73.2 ± 11.3, endrin-69.1 ± 6.4, p,p'DDD-79.6 ± 7.4, p,p'DDE-73.1 ± 10.4, and p,p'DDT-74.5 ± 5.4.

RESULTS AND DISCUSSION

Organic contaminants were generally low in all samples, whether in whole fish or fillets (Tables 1-3). No PCBs were detected in any samples, and pesticides were normally <10ppb and seldom >50ppb, with endosulfan, p,p'DDE, and delta BHC being detected most frequently. Whole fish and livers usually had a higher incidence of pesticides than fillets, i.e. 90% (n=73) of the whole fish, 80% (n=10) of the livers, and 32% (n=106) of the fillets had levels >1ppb for individual compounds. These results are consistent with the lipophilic character of pesticides as opposed to the water soluble nature of metals in general. No concentrations for whole fish, livers, or fillets exceeded any Food and Drug Administration (FDA) action levels (FDA 1992) or international limits Nauen (1983) for fish (Tables 1-3).

Although Charleston Harbor and Winyah Bay had most of the pesticide maxima, these concentrations were not high compared to some national levels. The National Oceanic and Atmospheric Administration (NOAA) reported total DDT (tDDT) maxima up to 17.4ppm in muscle tissue from a California fish (Mearns et al. 1988), while the tDDT maximum from this study was 82.4ppb for a seatrout fillet (Table 1).

On a regional basis, however, some of the pesticide maxima from this study are relatively high. Pesticide maxima for the Atlantic croaker (*Micropogonias undulatus*) from the Gulf of Mexico were lower in every case except for DDD, e.g. endosulfan and DDT were 58.0ppb and 82.4ppb (Tables 2-3) vs. 1.7ppb and 24.2ppb for the Gulf (Summers et al. 1993). These and the national data may reflect actual differences in levels of environmental contamination or in life histories of the respective organisms.

Metal concentrations were generally low in all three species at each location, but some individual maxima were relatively high (Table 4). Arsenic concentrations, for instance, were

Table 1. Pesticide Concentrations (Ranges and Means) in Red Drum (ppb wet weight).

Pesticide	Winyah Bay	Charleston Harbor	St. Helena Sound	Port Royal Sound	Criterion[1] ppb
alpha BHC	ND[2] (7)	ND (33)	ND (5)	ND (2)	-
beta BHC	ND-6.67 0.95±2.33(7)	ND - 19.1 2.20±4.86(33)	ND (5)	ND (2)	-
gamma BHC	ND (7)	ND - 7.96 0.24±1.37(33)	ND (5)	ND (2)	200
delta BHC	ND - 31.9 4.56±11.16(7)	ND - 187 23.9±43.6(33)	ND (5)	ND (2)	-
Heptachlor	ND (7)	ND (33)	ND (5)	ND (2)	300
Heptachlor Epoxide	ND (7)	ND - 11.9 0.36±2.04(33)	ND - 4.60 0.92±1.84(5)	ND (2)	300
alpha Chlordane	ND (7)	ND - 19.5 0.59±3.34(33)	ND (5)	ND (2)	300
gamma Chlordane	ND - 39.1 5.59±13.68(7)	ND (33)	ND (5)	ND (2)	300
Aldrin	ND (7)	ND (33)	ND - 5.73 2.16±2.65(5)	ND, 16.8	300
Dieldrin	ND (7)	ND (33)	ND (5)	ND (2)	300
Endrin	ND - 21.7 5.61±8.94(7)	ND - 21.4 0.65±3.67(33)	ND (5)	ND (2)	300
Endosulfan I	ND - 21.3 5.93±9.38(7)	ND - 6.27 0.60±1.56(33)	1.88 - 9.10 3.51±2.80(5)	ND (2)	-
Endosulfan II	ND - 36.6 5.23±12.81(7)	ND - 20.0 1.18±3.87(33)	ND (5)	ND (2)	-
p,p'DDD	ND (7)	ND(33)	ND(5)	ND (2)	5000
p,p'DDE	ND - 34.9 7.80±13.00(7)	ND - 70.2 5.00±13.68(33)	ND - 70.2 1.40±2.81(5)	ND (2)	5000
p,p'DDT	ND - 58.7 8.39±20.54(7)	ND - 12.1 0.37±2.07(33)	ND - 5.10 1.02±2.04(5)	ND (2)	5000

1. Limits from FDA (1992) and Nauen (1983).
2. ND = <1.0ppb.

up to 22.5ppm (flounder fillet), but the mean for all species was 1.88ppm (whole fish and fillets), which falls below the international limit of 2.0ppm for arsenic (Nauen 1983). The means for Port Royal Sound seatrout and both Winyah Bay and Charleston Harbor flounder were above the international limit (Table 4). Compared to Gulf croaker with an arsenic maximum of 2.1ppm (Summers et al. 1993), the above maximum and mean appear quite high. These levels may not be unusually high for the area, since Winger et al. (1990) reported arsenic means of 4.83ppm and 2.33ppm for sea catfish (*Aurius felis*) and Atlantic croaker respectively in the lower Savannah River. In addition, the form of arsenic most common in

Table 2. Pesticide Concentrations (Ranges and Means) in Seatrout (ppb wet weight).

Pesticide	Winyah Bay	Charleston Harbor	St. Helena Sound	Port Royal Sound	Criterion[1] ppb
alpha BHC	ND[2] (19)	ND (20)	ND (10)	ND (9)	-
beta BHC	ND (19)	ND (20)	ND (10)	ND (9)	-
gamma BHC	ND - 4.55 0.35±1.09(19)	ND - 17.0 1.25±3.81(20)	ND - 18.0 3.64±6.18(10)	ND (9)	200
delta BHC	ND (19)	ND - 43.6 3.15±10.19(20)	ND (10)	ND (9)	-
Heptachlor	ND (19)	ND (20)	ND (10)	ND (9)	300
Heptachlor Epoxide	ND - 10.7 0.56±2.39(19)	ND - 11.0 1.32±3.34(20)	ND - 6.23 0.92±1.97(10)	ND - 9.29 1.03±2.92(9)	300
alpha Chlordane	ND (19)	ND - 7.01 0.35±1.53(20)	ND - 13.3 2.70±4.89(10)	ND - 10.1 1.12±3.17(9)	300
gamma Chlordane	ND (19)	ND - 4.64 0.38±1.17(20)	ND - 5.02 0.89±1.79(10)	ND - 12.4 1.38±3.90(9)	300
Aldrin	ND - 12.4 1.67±3.91(19)	ND - 12.4 1.72±4.09(20)	ND - 6.10 0.61±1.83(10)	ND - 7.33 1.34±2.58(9)	300
Dieldrin	ND (19)	ND - 15.6 0.78±3.40(20)	ND (10)	ND (9)	300
Endrin	ND - 49.9 2.68±11.13(19)	ND (20)	ND (10)	ND (9)	300
Endosulfan I	ND - 17.0 1.73±5.04(19)	ND - 34.6 5.46±10.01(20)	ND - 21.3 2.57±6.38(10)	ND (9)	-
Endosulfan II	ND (19)	ND - 41.5 2.08±9.04(20)	ND (10)	ND (9)	-
p,p'DDD	ND 2.94 0.16±0.66(19)	ND (20)	ND - 8.66 0.87±2.60(10)	ND (9)	5000
p,p'DDE	ND - 8.54 1.94±5.49(19)	ND - 56.3 6.87±15.17(20)	ND - 14.4 1.64±4.29(10)	ND (9)	5000
p,p'DDT	ND - 82.4 4.34±18.44	ND (20)	ND (10)	ND - 70.2 14.6±22.0(9)	5000

1. Limits from FDA (1992) and Nauen (1983).
2. ND = <1.0ppb.

the marine biota, arsenobetaine, is the least toxic to marine organisms and humans (Eisler 1988).

Lead concentrations were low overall, but the mean of 0.577ppm for all samples and several individual means (Table 4) exceeded the international limit of 0.5ppm (Nauen 1983). The maximum of 9.68ppm in a Winyah Bay red drum (whole fish) was much higher than the maximum in Gulf croaker of 0.3ppm (Summers et al. 1993). Since 71% (n=127) of all

Table 3. Pesticide Concentrations (Ranges and Means) in Flounder (ppb wet weight).

Pesticide	Winyah Bay	Charleston Harbor	St. Helena Sound	Port Royal Sound	Criterion[1] ppb
alpha BHC	ND[2] (33)	ND (26)	ND (13)	ND (6)	-
beta BHC	ND - 4.04 0.28±0.91(33)	ND - 1.70 0.65±3.26(26)	ND (13)	ND (6)	-
gamma BHC	ND - 1.40 0.073±0.29(33)	ND - 9.15 0.65±2.26(26)	ND (13)	ND (6	200
delta BHC	ND - 5.70 0.17±0.97(33)	ND - 6.37 0.35±1.31(26)	ND - 9.62 0.76±2.56(13)	ND (6)	-
Heptachlor	ND - 7.74 0.24±1.33(33)	ND - 12.8 0.67±2.58(26)	ND (13)	ND (6)	300
Heptachlor Epoxide	ND - 2.76 0.084±0.473(33)	ND - 4.95 0.19±0.95(26)	ND (13)	ND (6)	300
alpha Chlordane	ND (33)	ND - 22.7 1.60±4.98(26)	ND (13)	ND (6)	300
gamma Chlordane	ND (33)	ND - 17.8 1.89±4.32(26)	ND - 9.57 0.74±2.55(13)	ND (6)	300
Aldrin	ND - 8.48 0.54±1.84(33)	ND - 20.2 0.78±3.89(26)	ND - 10.4 1.21±3.01(13)	ND (6)	300
Dieldrin	ND (33)	ND (26)	ND (13)	ND (6)	300
Endrin	ND - 4.72 0.14±0.81(33)	ND (26)	ND (13)	ND (6)	300
Endosulfan I	ND - 2.56 0.19±0.61(33)	ND - 58.0 3.87±12.56(26)	ND - 44.2 8.4±12.9(13)	ND - 34.1 5.68±12.71(6)	-
Endosulfan II	ND - 6.74 0.20±1.16(33)	ND - 21.6 1.00±4.20(26)	ND (13)	ND (6)	-
p,p'DDD	ND - 10.3 0.52±2.08(33)	ND (26)	ND (13)	ND (6)	5000
p,p'DDE	ND - 11.6 1.01±2.90(33)	ND - 30.9 4.07±8.91(26))	ND - 10.2 3.48±8.53(13)	ND - 18.6 3.10±6.93(6)	5000
p,p'DDT	ND - 70.6 2.73±12.14(33)	ND (26)	ND (13)	ND (6)	5000

1. Limits from FDA (1992) and Nauen (1983).
2. ND = <1.0ppb.

samples was <0.5ppm, however, elevated lead levels were not present in most samples.

Cadmium and mercury levels were consistently low throughout the sampling area and with respect to species (Table 4). Mean concentrations for all fillets and whole fish were 86.2ppb (cadmium) and 56.8ppb (mercury), with 70.7% (n=164) and 69.3% (n=88) of the samples being <25ppb cadmium and mercury respectively. Most of the cadmium and mercury detected was in whole fish samples, since 75.6% and 100% of the fillets were below detection

Table 4. Metal Concentrations (Ranges and Means) in Red Drum (ppm[1] wet weight)

Metal	Winyah Bay	Charleston Harbor	St. Helena Sound	Port Royal Sound	Criterion (ppm[1])
Cadmium	ND[3]-0.42	ND-0.41	ND(5)[4]	ND(1)	0.5
	0.04±0.10(35)	0.16±0.14(16)			
Lead	ND-9.68	ND-4.46	ND(5)	1.93(1)	0.5
	0.80±2.20(35)	1.28±1.21(16)			
Mercury	51-96	ND-66	ND(2)	---	1000
	75±16(4)	9.8±22.6(16)			
Arsenic	ND-9.24	ND-7.63	ND(5)	9.56(1)	2.0
	1.39±2.66(22)	1.44±1.65(25)			
Chromium	ND-0.47	ND-1.68	ND(5)	ND(1)	1.0
	0.12±0.26(34)	0.16±0.42(15)			
Nickel	ND-1.08	ND-2.85	ND(5)	0.93(1)	---
	0.24±0.29(35)	1.02±0.76(15)			
Copper	ND-5.60	ND-52.9	ND(5)	0.40(1)	15
	0.86±1.25(22)	9.19±18.05(16)			
Metal Concentrations (Ranges and Means) in Seatrout (ppm[1] wet weight)					
Cadmium	ND[3]-0.50	ND(12)[4]	ND-0.50	ND-0.46	0.5
	0.16±0.20(12)		0.07±0.16(8)	0.08±0.16(9)	
Lead	ND-2.18	ND-3.65	ND-0.680	ND-1.93	0.5
	0.33±0.62(11)	0.68±1.05(12)	0.20±0.22(8)	0.83±0.69(10)	
Mercury	ND-687	ND-258	ND-88.0	ND(2)	1000
	182±244(6)	88.2±156.5(11)	44.0±41.0(6)		
Arsenic	ND-3.10	ND-0.73	ND(4)	ND-13.3	2.0
	0.46±0.92(11)	0.22±0.52(12)		4.19±4.23(11)	
Chromium	ND-0.58	ND-3.46)	ND(8)	ND-1.29	1.0
	0.07±0.17(11)	0.29±0.96(12		0.25±0.48(9)	
Nickel	ND-12.6	ND-7.20	ND-5.13	ND-1.00	---
	1.95±3.72(11)	0.61±1.99(12)	0.64±1.70(8)	0.50±0.34	
Copper	ND-16.2	ND-19.0	ND-0.24	ND-12.9	15
	2.33±4.49(11)	2.63±5.93(12)	0.03±0.08(7)	2.9±3.9(10)	
Metal Concentrations (Ranges and Means) in Flounder (ppm[1] wet weight)					
Cadmium	ND[3]-0.78	ND-0.71	ND-0.71	ND(3)[4]	0.5
	0.16±0.28(28)	0.07±0.18(20)	0.09±0.22(16)		
Lead	ND-2.30	ND-5.78	ND-1.23	0.24-0.30	0.5
	0.16±0.42(35)	0.59±1.36(20)	0.28±0.38(16)	0.43±0.23(3)	
Mercury	ND-246	ND-730	ND-26.0	2.1-210	1000
	82±116(3)	81.4±171.8(20)	5.8±9.4(11)	74.7±95.8(3)	
Arsenic	ND-11.1	ND-22.5	ND(3)	ND(2)	2.0
	3.15±3.08(29)	4.80±6.26(15)			
Chromium	ND-2.28	ND(20)	ND-0.10	ND(3)	1.0
	0.29±0.55(28)		0.01±0.024(16)		
Nickel	ND-1.08	ND-6.30	ND-8.21	ND-5.3	---
	0.18±0.28(31)	0.42±1.43(20)	3.66±3.81(16)	1.77±2.50(3)	
Copper	ND-10.0	ND-22.2	ND-5.35	ND,2.95	15
	1.13±2.53(30)	1.47±4.99(20)	1.89±1.71(14)		

[1] Mercury in ppb.
[2] Limits from FDA (1992) and Nauen (1983).
[3] ND=<0.010 ppm (CD), <0.050 ppm (Pb), <0.90 ppb (Hg), <0.010 ppm (As), <0.060 ppm (Cr and Ni) and <0.040 ppm(Cu).
[4] Numbers of samples in parentheses.

limits for cadmium (<10ppb) and mercury (<0.9ppb) respectively. The overall means above and all individual means (Table 4) are well below both FDA and international limits.

Copper, nickel, and chromium maxima for the target species were high relative to corresponding Gulf croaker maxima, i.e. 52.9ppm, 12.6ppm, and 3.46ppm (Table 4) vs. 5.3ppm, 0.3ppm, and 0.3ppm (Summers et al. 1993). The actual means of 2.23ppm (copper),

0.831ppm (nickel), and 0.112ppm (chromium) did not exceed either the FDA or international limits. with 61.3% (n=150) of the copper being <0.5ppm and 58.5% (n=164) of the nickel and 91.4% (n=151) of the chromium being <0.10ppm.

In summary, the South Carolina estuarine finfish in this study (red drum, seatrout, and flounder) overall were not heavily contaminated from a human health perspective with the targeted contaminants. Although some means exceeded international limits, means were generally low when compared to earlier data or FDA action levels. Apparently organic and metallic contaminants were not greatly bioconcentrated, which was an encouraging finding when considering the level of coastal development over the past two decades. What the actual impacts on the target species are in terms of toxicity and sublethal effects is unknown and is a topic for future studies.

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