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Contaminants in Fishes from Johnston Atoll

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Abstract. This study examined the distribution of military-industrial contaminants accumulating in coral reef fishes in the lagoon of Johnston Atoll, Pacific Ocean. This atoll was a major military base involved in nuclear and chemical weapons as well as being a depot, transient airfield and harbor since the 1930's. The base was closed and abandoned in 2003. Fishes of different trophic levels were sampled from locations throughout the atoll. Contaminants of concern included radionuclides, heavy metals (antimony, arsenic, barium, cadmium, chromium, copper, lead, mercury and zinc) and organic contaminants including; polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs), herbicides, dioxins and furans (PCDD/PCDFs). The northwest section of Johnston Island, the largest island in the atoll, was the area with the highest level and variety of contaminants in fishes and sediments. This was near the site of the open burn pit and trash dump, fire training and explosives detonation areas, and the former storage site of Herbicide (Agent) Orange.

Keywords: Central Pacific Ocean, PCBs, PAHs, dioxin, Agent Orange, weapons grade plutonium

Introduction

Johnston is a remote Central Pacific Atoll. The nearest landfall is French Frigate Shoals, 804 km (500 miles) north. The atoll is about 1,287 km (800 miles) southwest of Honolulu, Hawaii and 1,440 km (900 miles) north of the Line Islands of Kiribati. Johnston Atoll (JA) came under military control in 1934 and remained so until 2003. Its use varied during those years but included extensive use as a refueling site, atmospheric nuclear testing, master LORAN station for the Pacific, storage site for unused herbicide orange (agent orange) and chemical weapons, and the incineration of chemical weapons in the Johnston Atoll Chemical Ammunition Disposal System (JACADS) (Lobel and Lobel 2008).

Many of the activities mentioned above, as well as infrastructure needed to support a military and civilian workforce of up to 2000 people, contributed to soil and sediment contamination within the atoll (Lobel and Kerr 2002). Polycyclic aromatic hydrocarbon, petroleum hydrocarbon and metal contamination was associated with refuse burning, fire training operations and fuel storage. Organochlorine contamination due to leakage from discarded electrical equipment, transformers, polychlorinated biphenyl (PCB) contaminated fuel and herbicide orange (HO) were the main contaminants of potential concern (COPC) due to accumulation in top predators and potential toxicity. HO contains two active ingredients, the n-butyl ester of 2,4- dichlorophenoxyacetic acid (2,4-D) and 2,4,5-trichlorophenoxyacetic acid (2,4,5-T), as well as the contaminant 2,3,7,8-tetrachlorodibenzo-p-dioxin (TCDD).

Polychlorinated biphenyls were found mainly in two areas of the lagoon, the west end of Sand Island and in the Navy Pier (tank 49 lagoon) area on Johnston Island (JI), the main island within the atoll. Polychlorinated dibenzo-p-dioxin and dibenzofuran (PCDD/PCDF) contamination in soil and nearshore sediments of the northwest corner of JI was caused by leaking storage drums containing HO (Lobel and Kerr 2002).

Of the four aborted nuclear tests that occurred on JI, two would have contributed to the dispersal of radionuclides into the lagoon. Most of the debris and residual plutonium from the STARFISH event, aborted at 30,000 feet, landed in the water surrounding JI and on adjacent Sand Island. The BLUEGILL PRIME event scattered radioactive material primarily downwind of the launch emplacement. The single residual contaminant was Weapons Grade Plutonium (WGP), which consists of five isotopes of plutonium (238, 239, 240, 241, and 242) and americium-241.

This review summarizes contaminant data in aquatic biota from multiple studies/sampling events sponsored by different agencies. Management responsibility for JA is shared among several agencies including the US Air Force, US Army, US Coast Guard, US Fish and Wildlife Service and the Defense Threat Reduction Agency thus, an integrated approach to ecosystem management including the need for long-term monitoring and potential cleanup is necessary.

Materials and Methods

Samples were collected from sites with known sediment contamination as well as sites with no sediment contamination for comparison (Lobel and Kerr 2002). The main concerns were PAHs, PCDD/PCDFs and metals in fishes from the NW corner of JI and down wind from JACADS, PCBs from the Tank 49 lagoon and Sand Island, radionuclides surrounding JI and Sand Island. Fish sampled from Kaneohe Bay, Oahu, Hawaii served as a reference site for radionuclides.

Sample species were chosen based on food habits or potential for accumulating contaminants, relative abundance and distribution within the atoll. Species sampled included parrotfishes *Chlorurus sordidus, Scarus psittacus, S. rubroviolaceous*; surgeonfishes *Ctenochaetus strigosus* and *Acanthurus triostegus*; the damselfish *Abudefduf sordidus*; goatfishes *Mulloidichthys flavolineatus Parupeneus multifasciatus, P. trifasciatus;* the triggerfish *Rhinecanthus aculeatus;* and the squirrelfish *Sargocentron tiere.*

Fish samples were collected by spear. Individual fish were placed in plastic bags after capture. Each sample location was mapped using GPS then plotted on nautical charts and high definition aerial photographs. Samples were chilled on ice immediately upon collection. Fish were identified to species, weighed and measured (SL). Samples were again cooled to 4°C prior to shipment on dry ice. Quality control samples consisted of approximately 10% of the total samples sent for analysis. Splits, duplicate samples, matrix spikes as well as field, trip, and laboratory blanks were used to determine the precision and accuracy of the analytical data. All analyses were completed by government contract laboratories using US Environmental Protection Agency (EPA) approved methods (USEPA 1993, 1994,1996, 1999; SW-846 method number in parentheses; http://www.epa.gov/sw-846/main.htm). Samples were extracted and separated using methods 3500, 3540(C) and 8000. Chlorinated herbicides were measured with gas chromatography (GC) using methylation (8150). Individual congeners of polychlorinated biphenyls (PCBs) were measured using high resolution GC and low resolution mass spectrophotometry (HRGC/LRMS; 8082A, modification 680 and EPA Method 1668). Total PCBs were taken as the sum of all mono- to decachlorinated congeners. Polychlorinated dibenzodioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) were measured using HRGC/HRMS (8290). Toxicity equivalents (TEQ) for 2,3,7,8 tetrachlorodibenzo-p-dioxin were calculated using fish derived toxicity equivalency

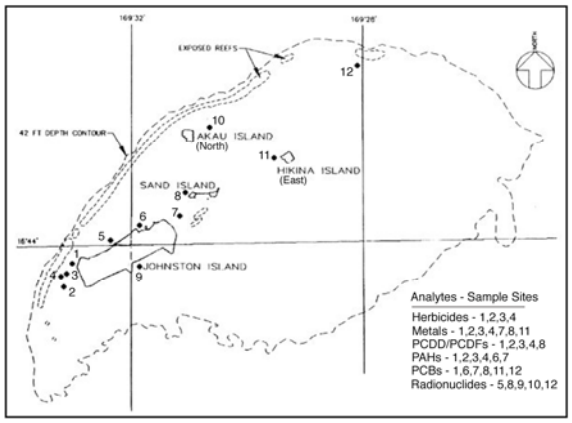

Figure 1. Biota sampling sites within Johnston Atoll. 1) offshore of the former Herbicide Orange (HO) storage area, 2) west camera stand, 3) across the channel from the HO site, 4) inner reef area, 5) offshore of the radiation control area (RCA), 6) tank 49 lagoon area, 7) Buoy 14, 8) west end of Sand Island, 9) south side of Johnston Island, 10) Blue Hole, 11) East Island, 12) Donovan's Reef.

factors (TEF) for 17 2,3,7,8 substituted congeners (Van den Berg et al. 1998). Polycyclic aromatic hydrocarbons (PAHs) were measured with GC/MS (8270). Barium, cadmium, chromium, copper, lead, and zinc were measured using atomic absorption (AA) with direct aspiration (7080, 7130, 7190, 7210, 7420, 7950) while antimony and arsenic used a borohydride reduction (7062). Mercury was measured using the cold vapor technique (7471A).

Radionuclide analysis was conducted by Oak Ridge National Laboratory (ORNL), Grand Junction, Colorado. Whole fish bodies without viscera and the viscera were analyzed separately by alpha spectrometry for ^{241}americium, ^{244}curium, ^{238}plutonium, $^{239+240}$plutonium, and ^{242}plutonium using ORNL procedures PU242 and RC-19 R06.

Results

Multiple sampling events occurred between 1995 and 2003 focusing on different areas of the atoll and differing contaminants (Fig.1).

In total, 94 reef fish samples were analyzed for PCBs, 80 for PCDD/PCDFs and metals, 55 for the herbicides 2,4-D and 2,4,5-T, 65 for PAHs, and 82 for radionuclides.

The herbicides 2,4 D and 2,4,5 T were not detected in any fish tissues. Trace levels of PAHs were detected in fish tissues from sites 1,2,3,4 and

6 but all detections were below the limits of quantification.

The most toxic dioxin congener, 2,3,7,8 tetrachlorodibenzo-p-dioxin (TCDD) was detected in 35% of the 80 fish samples analyzed for PCDDs/PCDFs. Of these samples 79% (27 of 31) of the detections were from offshore of the former

Table 1. Distribution and concentration of A) organic constituents including percent lipids, B) metals and C) radionuclides in Johnston Atoll reef fish tissue and viscera by site. Sample sizes (N) are the number of individual fish analyzed with the number of species collected at each site shown in parentheses. Data are summarized as mean ±sd (range) wet weight (ww). Sample sites are shown in Fig. 1.

A – Organic concentrations (whole body, pg/g or ng/g ww)

Site	N	TCDD (pg/g)	TEQ (pg/g)	∑PCBs (ng/g)	Lipid (%)
1	31 (8)	6.3 ± 8.8 (0.6-40.5)	5.5 ± 8.4 (0-40.6)	35.0 ± 27.6 (6.8-77.5)	3.6 ± 1.4 (1.6-5.2)
2	13 (5)	ND	0.02 ± 0.03 (0-0.08)	4.1 (N=1)	3.8 ± 3.4 (0.7-11.2)
3	12 (6)	0.82	0.2 ± 0.7 (0-2.4)	NA	3.6 ± 3.1 (0.8-8.7)
4	6 (4)	0.4	0.1 ± 0.2 (0-0.5)	NA	4.5 ± 2.4 (1.4-7.8)
6	35 (5)	NA	NA	1432 ± 1538 (52.4-6789)	3.5 ± 1.8 (0.6-9.0)
7	18 (1)	ND	ND	36.6 ± 52.3 (3.9-230.0)	3.5 ± 2.0 (1.1-9.3)
8	18 (1)	ND	0.04 ± 0.03 (0-0.08)	753.5 ± 900 (29.7-3270)	3.9 ± 1.4 (1.8-7.1)
11	12 (1)	ND	0.0009	12.8 ± 9.2 (3.4-38.7)	4.0 ± 2.1 (1.7-8.0)
12	8 (1)	NA	NA	15.1 ± 31.0 (3.7-91.8)	2.7 ± 2.2 (0.6-6.0)

B – Metal concentrations (whole body, µg/g ww)

Site	N	As	Ba	Cd	Cr	Cu	Hg	Sb	Pb	Zn
1	31 (8)	2.0 ± 0.2 (1.8-2.3)	1.0 ± 0.3 (0.6-1.3)	0.04 ± 0.02 (0.03-0.08)	0.6 ± 0.5 (0.3-1.5)	0.6 ± 0.1 (0.5-0.7)	0.03 ± 0.01 (0.02-0.05)	0.01	0.5 ± 0.2 (0.2-0.9)	15.7 ± 4.3 (11.0-22.5)
2	13 (5)	2.1	13.2 ± 7.4 (1.0-19.4)	0.3	0.4	0.7	0.02	ND	0.07	15.0 ± 8.5 (5.8-34.2)
3	12 (6)	ND	13.7 ± 2.0 (12.4-16.0)	ND	ND	ND	0.05	NA	ND	15.1±10.10.1 (6.7-43.1)
4	6 (1)	ND	15.8	ND	ND	ND	ND	NA	ND	14.1± 6.9 (7.8-24.2)
7	6 (1)	2.0 ± 0.8 (1.3-3.4)	1.4 ± 0.6 (0.7-2.3)	0.08 ± 0.05 (0.03-0.2)	0.3 ± 0.1 (0.2-0.4)	0.6 ± 0.1 (0.5-0.7)	0.02 ± 0.001 (0.02-0.03)	ND	0.5 ± 0.3 (0.2-1.0)	19.1 ± 3.4 (12.5-22.1)
8	6 (1)	2.2 ± 0.5 (1.6-2.8)	0.8 ± 0.5 (0.4-1.8)	0.04 ± 0.01 (0.02-0.1)	0.4 ± 0.1 (0.4-0.6)	0.7 ± 0.2 (0.6-1.0)	0.08 ± 0.04 (0.03-0.1)	ND	1.7 ± 2.1 (0.6-6.0)	20.7 ± 5.1 (16.1-28.4)
11	6 (1)	2.6 ± 0.2 (2.3-2.9)	0.5 ± 0.2 (0.2-0.7)	0.1 ± 0.05 (0.05-0.2)	0.5 ± 0.3 (0.4-1.1)	0.7 ± 0.05 (0.7-0.8)	0.1 ± 0.1 (0.04-0.3)	ND	0.1 ± 0.05 (0.1-0.2)	18.5 ± 2.6 (15.0-21.6)

C – Radionuclide concentrations (pCi/kg ww)

		^{238}Pu (pCi/kg)		$^{239+240}$Pu (pCi/kg)		^{242}Pu (pCi/kg)		^{241}Am (pCi/kg)	
Site	N	Body	Viscera	Body	Viscera	Body	Viscera	Body	Viscera
5	25 (5)	0.2 ± 0.1 (0-0.4)	7.7 ± 13 (0-55)	2.9 ± 3 (0.4-10)	481 ± 805 (4.9-3816)	ND	3.0 ± 1 (1.5-4.3)	0.4 ± 05 (0-1.4)	77.9 ± 147 (0-654)
8	15 (3)	ND	3.7 ± 2 (0-6.6)	0.4 ± 0.04 (0-0.4)	26.4 ± 18 (0-60.1)	0.2	1.8	ND	11.4 ± 5 (4.5-20.2)
9	10 (1)	0.2	2.0	0.7 ± 0.3 (0-0.9)	13.6 ± 6 (0-28.7)	ND	1.4	ND	6.6 ± 2 (0-8.7)
10	10 (1)	0.3 ± 0.01 (0-0.3)	ND	ND	5.3 ± 2 (0-7.8)	ND	2.1 ± 0.5 (0-2.5)	0.3 ± 0.03 (0-0.3)	3.6 ± 1 (0-5.2)
12	25 (6)	0.5	4.2 ± 3 (0-8.6)	0.7 ± 0.5 (0-1.5)	6.3 ± 4 (0-10.1)	0.3	ND	0.5	5.3 ± 2 (0-7.4)
HI *	8 (2)	0.4 ± 0.2 (0-0.6)	4.2	ND	ND	0.2	2.6	ND	3.7

ND – none detected, NA – not analyzed, * Kaneohe Bay, Oahu, Hawaii

Table 2. Comparison of mean PCB, $^{239+240}$Pu, and metal concentrations in fishes from different Pacific locations. $^{239+240}$Pu activity in muscle only from JA fish was calculated based on tissue partitioning factors and muscle mass from Noshkin (1987). Muscle activity = eviscerated fish activity X muscle partition factor (0.045 for surgeonfish and 0.075 for goatfish); Muscle mass = total fish mass X 0.663; Muscle concentration = muscle activity (pCi)/muscle mass (kg).

Location	∑PCB ng/g	As µg/g	Cd µg/g	Cr µg/g	Cu µg/g	Pb µg/g	Zn µg/g	$^{239+240}$Pu pCi/kg muscle
Johnston[*]	630.3	1.9	0.07	0.5	2.6	0.7	16.1	0.05
Hawaii[*]	-	-	-	-	-	-	-	ND
Midway[#a]	392.5	-	-	-	-	-	-	-
Tern Is.[#b,c]	12,726	25	1.0	12.2	36.8	26.6	28.0	-
Disappearing Is.[#b,c]	393	25	ND	8.0	7.8	10.5	7.0	-
Marshall Is.[*d]	-	-	-	-	-	-	-	0.1
Bikini[*e]	-	-	-	-	-	-	-	0.35

[*] wet weight, [#] dry weight, [a] Hope et al. (1997), [b]Miao et al. (2001), [c]Miao et al. (2000), [d]Robison et al. (1997), [e]Robison et al. (1997).

HO site (1) (Table 1). TCDD was only detected in one other sample (squirrelfish, *Sargocentron tiere*) from site 3. Other PCDD/PCDF congeners and dioxin-like chemicals including mono-ortho and non-ortho PCBs (Zabel et al. 1995, van den Berg et al. 1998) used to calculate TCDD toxicity equivalents (TEQ) were detected in fishes from sites adjacent to the former HO site (2,3,4) as well as East (12) and Sand Islands (8).

Fish tissue concentrations of dioxin related compounds did not exceed environmental concentrations associated with risk to aquatic life. The TEQ in fish tissue was 40.6 pg/g while concentrations associated with low risk to aquatic life is 50 pg/g (US EPA 1993).

PCBs were detected in all fish tissues, but in much higher and more variable concentrations from Sand Island (8) and Tank 49 lagoon (6), sites with known sediment PCB contamination (Table 1). Mean fish PCB concentrations at sites with no known PCB contamination ranged from 12.8 to 36.6 ng/g compared to 1,432 and 753.5 ng/g at contaminated sites. Ecological benchmarks reported for PCB concentrations in whole fish tissue are 0.1, 0.5, and 50 µg/g (Swain and Holmes 1985, Beyer et al. 1996, BCMOELP 1998). PCB concentrations ranging from 2.0 to 6.8 µg/g at site 6 and 0.3 to 3.3 µg/g at site 8 exceed the lower benchmarks.

During the 2001 sampling event all sites sampled (1,7,8,11) contained detectable metals compared to earlier sampling events where only Zn, Ba, and occasionally Hg and Cu were detected. There were no consistent site differences in metal concentrations.

Radionuclide analyses of reef fish specimens revealed that 10% of fish samples and 56% of viscera samples had detectable levels of ^{241}Am activity (Table 1). ^{238}Pu was detected in 16 and 35% of tissues and viscera. $^{239+240}$Pu activity was measured in 32 and 72% of fish tissues and viscera respectively. ^{244}Cu was only detected in one sample respectively from the RCA (5) and Sand Island (8). ^{242}Pu was detected in 6 and 12% of tissues and viscera. Radionuclide activity in reef fishes was highest offshore of the RCA (5). Fewer detections were found in Kaneohe Bay, Hawaii samples although ^{241}Am, ^{238}Pu, and ^{242}Pu activities were similar. $^{239+240}$Pu and ^{244}Cu were not detected in Hawaiian tissue or viscera (Table 2).

Discussion

The objective of this analysis was to provide a summary of the majority of contaminant data currently available for marine fishes from Johnston Atoll. With the exception of radionuclide analysis, fish whole bodies were analyzed to evaluate exposure and bioaccumulation.

Contaminants in fish tissues were greatest at sites with known sediment contamination. Compared to other contaminants, PCBs accumulated to much higher levels in fish tissues than found in sediments. Mean PCBs in sediments from site 6 and 8 were 86.6 and 35.7 ng/g compared to an average of 1432 and 735 ng/g in fish tissues (Lobel and Kerr 2002). In contrast, mean TCDD (at site 1) concentrations in tissues (5.5 pg/g TEQ) were lower than average sediment concentrations (17 pg/g TEQ). These differences may be a function of different levels of contaminant localization.

Radionuclide activity in fishes from JA was detected but low compared to locations in the Marshall Islands where extensive nuclear testing

occurred (Robinson et al. 1997). Ingestion and uptake can still occur however, since the highest radionuclide activities were found within the viscera. Noshkin (1987) suggested that plutonium uptake is lower, or that plutonium is less bioavailable to JA fish since it was produced during an aborted test and is likely a different form compared to the nuclear generated plutonium found at Bikini, Enewetak and other Marshall Island locations.

Assessing impacts of chemical contaminants in coral reef environments is difficult due to a lack of baseline monitoring criteria as well as appropriate screening guidelines for risk assessment (Jameson et al. 1998). Since tropical organisms may not respond to xenobiotics in the same manner or at the same concentrations as temperate organisms (Johannes and Betzer 1975), it is important to both quantify and monitor specific stressors and ecological responses in order to understand concentrations in the field at which adverse effects are observed (Peters et al. 1997).

These data provide baseline information for future surveys and for comparisons of chemical contaminants in the fish biota from Johnston Atoll to fishes in other coral reef habitats. An expanded analysis of this data set will examine the pattern of contamination by species and trophic guild (Lobel and Lobel ms). Long term monitoring is needed since the implications of these contaminants on the reef biota are still being evaluated.

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