Investigation of Corexit® 9500 Dispersant in Gulf of Mexico Seafood Species

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Assessment of the human and environmental health impacts of the Deepwater Horizon oil spill was complicated by the unprecedented use of chemical oil dispersants to combat the spill. Federal and State public health agencies charged with ensuring the safety of seafood developed protocols for re-opening of oil-impacted areas closed to seafood harvesting. Methodologies were developed and implemented for monitoring and assessment of petrochemical and dispersant residues in seafood. In this report, methodology for the Corexit® 9500 dispersant indicator, sodium dioctylsulfosuccinate, was applied to laboratory (dispersant-exposed) and field samples of seafood species, confirming its low potential for bio-concentration and rapid elimination from edible tissues. Based on these data, those of the literature concerning the environmental fate and toxicity of Corexit® 9500, along with well documented related studies from other independent scientists, we conclude that to date there is no evidence to support a public health concern from consumption of Gulf of Mexico seafood harvested from re-opened areas.

The April 20, 2010 explosion and subsequent sinking of the Deepwater Horizon oil production platform (DWH) resulted in the largest oil spill in U.S. history. On April 29th, a Spill of National Significance was declared as roughly 53 thousand barrels of oil per day flowed into the Gulf of Mexico (GOM) (1). The U.S. Coast Guard estimated 4.9 million barrels of crude oil escaped before the damaged DWH wellhead was sealed on July 15, 2010 (2). Among the response strategies implemented to reduce the impact of oil on marine and coastal environments was the use of chemical oil dispersants. An estimated 1.1 million gallons of Corexit® 9500 dispersant was sprayed over the surface of oiled areas in the GOM and 800,000 gallons was injected directly into the oil spilling from the well head 1 mile beneath the surface (3). The unprecedented use of chemical dispersants elicited concerns for the health of responders, marine ecosystems, and the safety of seafood from impacted areas of the GOM.

Ingredients of Corexit® 9500 include propylene glycol, petroleum distillates hydrotreated light fraction, sodium dioctylsulfosuccinate, and sorbitan oleate emulsifying agents (4). These chemical compounds are used in many consumer products including use as a food additive and in personal care products, cosmetics, and household cleaning products (5). Investigation of these chemical compounds suggests that this dispersant poses little risk from consumption of GOM seafood. Propylene glycol (CAS 57-55-6) is a generally recognized as safe (GRAS) food additive (6), among other common uses. The solvent, petroleum distillates hydrotreated light fraction (CAS 64742-47-8), is a mixture of food-grade n-alkanes ranging from nonane to hexadecane (7). The major nonionic surfactants are ethoxylated sorbitan mono- and trioleates and sorbitan monooleate (CAS 1338-43-8, 9005-65-6, 9005-70-3) commonly used in cosmetics, toothpaste, and other consumer products. The major anionic surfactant, sodium dioctylsulfosuccinate (CAS 577-11-7), is a wetting agent in food, industrial, and cosmetic applications and a stool softener in clinical use (e.g., docusate). Nevertheless, public concern and an abundance of caution suggested that FDA and the U.S. National Oceanic and Atmospheric Administration (NOAA) further assess GOM seafood for potential contamination from the dispersant. Sodium dioctylsulfosuccinate (DOSS) was selected as the most appropriate indicator compound for Corexit® contamination in seafood due to its bioactivity, extremely low volatility, and potential to persist in the environment longer than other dispersant components (8,9).

A rapid extraction procedure and liquid chromatography tandem mass spectrometry (LC-MS/MS) method were developed to test for the presence of DOSS in seafood (9). Briefly, a 5-g portion of homogenized seafood is fortified with a deuterated DOSS standard and mixed with 25% aqueous acetonitrile. Magnesium sulfate (6 g) and sodium acetate (1.5 g) are added and the mixture shaken vigorously. The preparation is then centrifuged at 3000 x g and a portion of the acetonitrile layer is filtered through a 0.2 μ filter. The extract is then analyzed for DOSS by LC-MS/MS. The method, available in supporting online materials, was validated at the FDA Forensic Chemistry Center (FCC) and the NOAA Northwest Fisheries Science Center (NWFSC) using 38 samples of seafood matrices (finfish, crab, shrimp, and oyster) fortified with DOSS at levels ranging from 0.1 to 1.0 ppm. The average recovery was 92% with a RSD of 11%. The method limit of detection (LOD) in seafood ranged from 0.003 μg/g (FCC) to 0.015 μg/g (NWFSC) and the method limit of quantitation (LOQ) ranged from 0.010 μg/g (FCC) to 0.045 μg/g (NWFSC) in seafood.

To assess the seafood safety implications of dispersant contaminated seawater, controlled laboratory exposures of live Eastern oyster (*Crassostrea virginica*), blue crab (*Callinectes sapidus*), and red snapper (*Lutjanus campechanus*) to Corexit® 9500 were performed. Although dispersant concentrations in DWH application scenarios have been estimated to be approximately 30 μg/L (10), and dilution in ocean environments result in decreases of dispersant concentrations below detection levels within hours (11,12), an exposure concentration of 100 mg/L was used in this study to maximize the probability of detecting dispersant in the edible portions of test animals. This exposure level falls in the mid-range of experimental LC_{50} values for Corexit® 9500 determined for a variety of marine species (11).

Oysters, crabs, and fish specimens were collected from Mobile Bay, AL, coastal waters of Dauphin Island, AL, and in the GOM, respectively, and acclimated in aerated holding tanks at the FDA Gulf Coast Seafood Laboratory (GCSL), Dauphin Island, AL (13). Forty-six live oysters and 21 live crabs were placed in 100 L aerated seawater dosing tanks. Seawater salinity was 20 parts per thousand (ppt) and water temperature was 20^{o}C. Waterborne exposures of oysters and crabs to the dispersant were performed by weighing 10 g of Corexit® 9500 into a glass beaker, pouring the test article into the dosing tanks and stirring. Aeration of tank water facilitated the mixing of dispersant in the seawater tanks. Oysters and crabs were exposed to the dispersant test article for a period of 24 or 48 h. Two crab and 13 oyster mortalities were recorded during the exposures and subsequent depuration periods. Control oysters were retained in holding cages near the seawater intake pier at GCSL, and control crabs were retained in the holding tank.

Eight red snapper were placed in recirculating seawater dosing tanks. Exposures of fish to the dispersant were performed by weighing 96.12 g of Corexit® 9500 into a 2 L glass Erlenmeyer flask, mixing with approximately 1.5 L with seawater from the tank, and pouring the mixture into the dosing tank. The flask was rinsed 4 times with 1.5 L seawater to assure that all of the Corexit® 9500 was added. The final concentration of Corexit® 9500 in the dosing tank was 100 mg/L. Temperature, salinity, and dissolved oxygen averaged 30.4^{o}C, 22.7 ppt, and 5.7 mg/L, respectively, during the exposure period. Four mortalities were observed during the 24 h exposure period. Two control fish were retained in the flow-through seawater holding tank system.

After 24 h exposure, 9 crabs were removed for sample processing and analysis for DOSS in edible tissues. Crabs were placed in ice to immobilize, rinsed under cold tap water, and dissected for hepatic, pancreas and muscle tissues. Tissues were separately homogenized and stored at -80ºC in glass storage bottles with polytetrafluoroethylene (PTFE)-lined closures. Remaining live crabs were transferred to a flow-through seawater flume. At intervals of 24, 48, and 72 h of depuration, crabs were removed from the seawater flume and processed for analysis as described above.

Eight oysters were removed from the exposure water at 24 and 48 h after addition of dispersant and brushed thoroughly under running tap water. Oysters were shucked, and tissues homogenized and frozen at -80ºC in glass storage bottles with PTFE-lined closures. Remaining live oysters were transferred to a flow-through seawater flume with continuous seawater feed. Six oysters were sampled at 24 and 48 h of depuration, and 5 oysters at 72 h, and the tissues processed for analysis as described above.

Two red snapper were sampled at 24 h after dispersant addition. The remaining fish were transferred to a recirculating seawater tank for depuration. One fish was sampled at each of 24 and 48 h of depuration. Fish were euthanized in a 0.25 g/L tricaine methanesulfonate (MS-222) solution. Muscle tissue was dissected from each fish, homogenized, and frozen at -80ºC in glass storage bottles with PTFE-lined closures.

Equal portions of tissue homogenates of oyster, crab, and fish were shipped to the FDA-FCC and the NOAA-NWFSC laboratories for analyses of DOSS residues (9). Mean DOSS concentrations in oysters are presented in Table 1. DOSS was not detected in control oysters. In exposed oysters, levels were approximately 18 and 12 μg/g at 24 and 48 h, respectively. DOSS levels declined by >99% to 0.023 μg/g within 72 h.

Mean DOSS concentrations in crab muscle tissues are presented in Table 2. DOSS was not found in control samples. In crabs exposed to Corexit® 9500 for 24 h (no depuration), DOSS levels in muscle were approximately 0.9 μg/g. Levels declined to 0.023 μg/g (>97%) within 72 h of depuration.

DOSS concentrations in crab hepatopancreas are presented in Table 3. DOSS was not detected in control tissues. In crabs exposed to 100 mg/L Corexit® 9500 for 24 h, the mean DOSS concentration in hepatopancreas was approximately 11 μg/g, more than 10-fold higher than corresponding muscle tissue. Levels increased initially during the first 24 h of depuration, possibly a result of the re-distribution of residues during excretion. Levels subsequently declined to 2.4 μg/g (>95%) at 72 h of depuration.

Mean DOSS concentrations in muscle tissues of red snapper exposed to 100 mg/L Corexit® 9500 for 24 h, and depurated for 24 and 48 h, are presented in Table 4. DOSS was not found in control tissues. In the exposed animals, concentrations in muscle tissues were near or below the LOQ.

This seafood exposure study was designed to generate incurred DOSS residues to support method development and validation under a compressed time schedule. Nevertheless, it was possible to incorporate a limited sampling of depuration time points. The levels measured in exposed and depurated samples suggest that DOSS is not concentrated in the edible tissues of commercially relevant seafood species. Moreover, DOSS was rapidly eliminated from edible tissues following exposure. Our results with parent DOSS are consistent with a previous report using ^{14}C-labeled DOSS in rainbow trout fingerlings (14). In the latter study, total residue concentrations achieved steady state in whole blood and carcass (bioconcentration factors of 3.5 and 3.8, respectively) at 12 h of waterborne exposure, and were rapidly eliminated. DOSS and its metabolites were concentrated in bile. However, extrapolation of biliary data to edible tissues, other species, and routes of administration is unfounded.

Our laboratory findings are consistent with results obtained from GOM re-opening and surveillance samples collected from June through October 2010. A subset comprising 299 GOM fisheries re-opening samples from oil-impacted areas of State (Table S1) and Federal (Table S2) waters, collected from June through October, were examined for DOSS contamination. DOSS was not detected above the LOQ in 98% of the samples analyzed. Of 119 samples analyzed from State waters, 3 crustacean samples showed DOSS levels ranging from 0.011 to 0.013 μg/g, just above the FCC LOQ. Of 180 samples analyzed from Federal waters, 3 finfish samples contained DOSS ranging from 0.057 to 0.10 μg/g. During the same time period, NOAA surveillance samples collected from re-opened waters showed the highest levels of DOSS detected, ranging from 0.048 to 0.29 μg/g in 7 of 26 samples (Table S3). However, all 7 of the surveillance samples showing detectable levels of DOSS were collected as fillets whereas those surveillance samples collected as whole fish specimens were negative for DOSS, suggesting post-capture contamination during the filleting process. Contamination could result if cleaning agents containing DOSS are used in the processing environment. Of all of the samples tested to date, only a few showed trace amounts of dispersants residue and they were well below the safety threshold[1] of 100 parts per million for finfish and 500 parts per million for shrimp, crabs and oysters (15, 16).

In summary, laboratory exposures to 100 mg/L Corexit® 9500 were sufficient to generate incurred DOSS residues in the edible tissues of seafood species. However, little or no bio-concentration was evident. Moreover, incurred DOSS residues were rapidly eliminated after exposure. Exposure concentrations of dispersant at sea are predicted to be orders of magnitude lower. The vast majority of re-opening and surveillance samples tested negative for DOSS. The experimental and field data reported here, along with well documented related studies from other independent scientists, support FDA, NOAA, and EPA assessments that there is no evidence to date to support a public health concern from consumption of GOM seafood harvested from re-opened areas (17,18).

We acknowledge that public perception and concern about the quality and safety of Gulf of Mexico seafood persists. While the petrochemical and dispersant residues may not be a health threat, the public fear of potential toxic effects on the seafood may have economic and behavioral health consequences. Anxiety and reluctance to consume coastal seafood negatively impacts fishing industry production and may create local loss of income. Increased individual and community economic stress may contribute to poorer health and behavioral health outcomes in the costal communities. As Federal and State agencies ensure the safety of seafood in this region, they must also take positive steps to educate and reassure the public about the actual level of health risk to mitigate their anxiety and reinstate consumer confidence in Gulf Region seafood.

References

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Supplementary Online Material in Appendix 1
Methods
Tables S1 to S3

Table 1. Mean[a] DOSS concentrations in oysters exposed to 100 mg/L Corexit® 9500 for 24 and 48h and depurated for 24, 48, and 72h.

Sample	Exposure (h)	Depuration (h)	FCC (μg/g)[b]	NWFSC (μg/g)[c]
Control	0	0	< LOQ	< LOQ
O24-0	24	0	18 (1.0)	17 (2.0)
O48-0	48	0	12 (0.01)	12 (2.0)
O48-24	48	24	0.20 (0.004)	0.21 (0.0)
O48-48	48	48	0.047 (0.001)	0.052 (0.001)
O48-72	48	72	0.023 (0.001)	< LOQ

[a]Mean of two replicates. The range is shown in parentheses.
[b]FCC Limit of Quantitation (LOQ) = 0.010 μg/g DOSS.
[c]NWFSC LOQ = 0.045 μg/g DOSS.

Table 2. Mean[a] DOSS concentrations in crab muscle tissue exposed to 100 mg/L Corexit® 9500 for 24 h and depurated for 24, 48, and 72h.

Sample	Exposure (h)	Depuration (h)	FCC (μg/g)[b]	NWFSC (μg/g)[c]
Control	0	0	< LOQ	< LOQ
CM24-0	24	0	0.91 (0.03)	0.91 (0.07)
CM24-24	24	24	0.49 (0.01)	0.58 (0.05)
CM24-48	24	48	0.14 (0.003)	0.19 (0.06)
CM24-72	24	72	0.023 (0.001)	< LOQ

[a]Mean of two replicates. The range is shown in parentheses.
[b]FCC Limit of Quantitation (LOQ) = 0.010 μg/g DOSS.
[c]NWFSC LOQ = 0.045 μg/g DOSS.

Table 3. Mean[a] DOSS concentrations in crab liver and pancreas tissue exposed to 100 mg/L Corexit® 9500 for 24h and depurated for 24, 48, and 72h.

Sample	Exposure (h)	Depuration (h)	FCC (μg/g)[b]	NWFSC (μg/g)[c]
Control	0	0	< LOQ	< LOQ
CH24-0	24	0	11 (0.8)[d]	11 (0.0)
CH24-24	24	24	54[e]	59 (4.0)
CH24-48	24	48	39[e]	45 (2.0)
CH24-72	24	72	2.4[e]	2.9 (0.0)

[a]Mean of two replicates. The range is shown in parentheses.
[b]FCC Limit of Quantitation (LOQ) = 0.3 μg/g DOSS.
[c]NWFSC LOQ = 0.045 μg/g DOSS.
[d]Range for 3 determinations.
[e]Only a single preparation was evaluated.

Table 4. Mean[a] DOSS concentrations in red snapper muscle tissue exposed to 100 mg/L Corexit® 9500 for 24h and depurated for 24 and 48h.

Sample	Exposure (h)	Depuration (h)	FCC (µg/g)[b]	NWFSC (µg/g)[c]
Control	0	0	< LOQ	< LOQ
Control	0	0	< LOQ	< LOQ
RS24-0	24	0	0.017 (0.004)	< LOQ
RS24-0	24	0	0.013 (0.002)	< LOQ
RS24-24	24	24	< LOQ	< LOQ
RS24-48	24	48	< LOQ	< LOQ

[a]Mean of two replicates. The range is shown in parentheses.

[b]FCC Limit of Quantitation (LOQ) = 0.010 µg/g DOSS.

[c]NWFSC LOQ = 0.045 µg/g DOSS.

Appendix 1

Supporting Online Material for

Investigation of Corexit® 9500 Dispersant in Gulf of Mexico Seafood Species

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Title: **Investigation of Corexit® 9500 Dispersant in Gulf of Mexico Seafood Species**

Supplemental Material

Supplemental Data

The internet link http://www.fda.gov/downloads/ScienceResearch/FieldScience/UCM231510.pdf shows the analytical method for determination of sodium dioctylsulfosuccinate (DOSS) in seafood, including method performance and validation criteria.

Methods

Crab and Oyster Sample Collection

Blue crab (*Callinectes sapidus*) were collected from the south shoreline of Dauphin Island, AL, placed in baskets and transported to the GCSL. Crabs were transferred to 1,287 L recirculating aerated holding tanks (Red Ewald, Inc.; Karnes City, TX) supplied with seawater pumped from the laboratory seawater intake pier.

Eastern oyster (*Crassostrea virginica*) were collected from Mobile Bay, AL shellfish beds at the mouth of Fowl River and transported to the FDA Gulf Coast Seafood Laboratory (GCSL), Dauphin Island, AL. At the GCSL, oysters were transferred to wire-mesh cages and placed on the sea bottom near the laboratory seawater intake pier.

Crab and Oyster Exposure, Depuration, and Sample Preparation

Seawater exposure of crabs to the dispersant test article was performed by weighing 10 g of Corexit® 9500 into a glass beaker, pouring the article into the dosing tank and stirring. The glass beaker was placed on the bottom of the tank. Air bubbles generated by the air compressor connected to the air dispersion stones aided in mixing and dissolution of the dispersant in the seawater tank. Final dispersant test article concentration was 10 g Corexit® 9500 in 100 L seawater (100 ppm). Crabs were exposed to the dispersant test article for a period of 24 h. Control crabs were retained in the 1,287 L holding tank.

After 24 h exposure, nine crabs were removed for sample processing and analysis for dispersant constituent(s) in edible tissues. Crab sample processing included placing the live crabs in ice to immobilize, rinsing under cold tap water, removal of the carapace, removal of the liver and pancreas, removal of the muscle tissue, homogenization of the liver and pancreas and muscle tissue, and freezing at -80ºC in 60 or 125 ml glass storage bottles with PTFE lined closures (IChem; Thermo Fisher Scientific, Rockwood, TN). The remaining crabs were transferred to a flow-through seawater flume with continuous feed from the seawater intake pier. At intervals of 24, 48, and 72 h of depuration, crabs were removed from the seawater flume and processed for analysis as described above.

Oyster exposure to 100 ppm Corexit® 9500 was conducted with 46 oysters using the same procedures as described above for crabs. After 24 and 48 h exposed oysters were removed from the exposure tank, brushed thoroughly under running tap water, shucked, homogenized, and frozen at -80ºC in 60 or 125 ml glass storage bottles with PTFE-lined closures. Remaining oysters were transferred to a flow-through seawater flume with continuous feed from the seawater intake pier. At 24, 48, and 72 h of depuration, oysters were removed from the seawater flume and processed for analysis as described above.

Red Snapper Exposure, Depuration, and Sample Preparation

Red snapper (*Lutjanus campechanus*) were harvested from the Gulf of Mexico near oil rigs located in Mobile Oil and Gas Lease Blocks 914 and 959 using the hook and line fishing method. Each snapper had its swim bladder deflated immediately after landing on the boat by inserting a 16 gauge hypodermic needle through the skin and into the bladder. The fish were transferred to 208 L plastic drums containing approximately 150 L aerated seawater.

At the laboratory, 11 red snapper from the aerated drums were transferred to one of two 1,287 L flow-through holding tanks supplied with seawater pumped from the laboratory seawater intake pier. The fish were allowed to acclimate in the tanks for 5 days. Overall fish health was monitored daily. Temperature, salinity, and dissolved oxygen in these tanks averaged 29.7^{o}C, 23.4 ppt, and 5.1 mg/L, respectively. Three fish died during the acclimation period.

After acclimation, the eight remaining live red snapper were transferred to a 1,287 L recirculating holding tank supplied with seawater pumped from the laboratory seawater intake pier. The total volume of seawater in the recirculating tank system was 961.2 L.

Once the fish had acclimated, 96.12 g of Corexit® 9500 was weighed into a 2-L Erlenmeyer flask. The flask was filled to approximately 1.5 L with seawater from the tank and swirled to mix. The Corexit® 9500/seawater solution was then gently poured into the recirculating seawater tank system. This step was repeated 4 times to assure that all of the Corexit® 9500 was added. The final concentration of Corexit® 9500 in the recirculating tank was 100 ppm. Temperature, salinity, and dissolved oxygen in this tank averaged 30.4^{o}C, 22.7 ppt, and 5.7 mg/L, respectively, during the 24-h exposure period.

Since eight red snapper were exposed to 100 ppm Corexit® 9500 in seawater, it was determined that two fish would be sampled at 24 h exposure. The remaining six fish would be transferred to a second recirculating seawater tank containing fresh seawater and allowed to depurate. Two fish would be sampled at 24, 48, and 72 h, respectively. However, four of the eight red snapper died at approximately 8, 8.75, 11.25, and 11.5 h of exposure, and were excluded from the study. The remaining four fish survived the 24 h exposure to 100 ppm Corexit® 9500 in seawater.

Two fish were sampled at the 24 h exposure period and two fish were transferred to fresh seawater for depuration. One fish was sampled at 24 h depuration and the final fish was sampled at 48 h depuration. Two control (unexposed) fish harvested separately were sampled after 3 days of acclimation in the flow-through seawater tank system.

All red snapper were euthanized by submersion in a 0.25 g/L MS-222 solution and processed as follows. Muscle tissue was dissected from each fish, homogenized (separately) in blender jars placed on a blender base, and frozen at -80ºC in 60 or 125 ml glass storage bottles with PTFE-lined closures.

Sample Shipment

All samples were split and equal portions were shipped (frozen on dry ice) via overnight courier to the FDA Forensic Chemistry Center (FCC) and the NOAA Northwest Fisheries Science Center (NWFSC) for analyses of DOSS residues. The samples were utilized for LC-MS/MS method development for DOSS as an indicator of Corexit® 9500 contamination in GOM seafood species.

Supplemental Tables

Table S1. Levels of DOSS in edible seafood tissue sample composites collected for re-opening of State harvest waters[a].

State	Collection Date	Sample Type	DOSS (µg/g)
LA	7/14/2010	Spotted sea trout	< LOD[b]
LA	7/14/2010	Black drum/Spotted seatrout	< LOD
LA	7/14-15/2010	Spotted seatrout/Southern kingfish/Gafftopsail catfish	< LOD
LA	7/15/2010	Gafftopsail catfish	< LOD
LA	7/15; 7/18/2010	Spotted seatrout/Atlantic croaker/Sand seatrout	< LOD
LA	7/18-19/2010	Spotted sea trout	< LOD
LA	7/14; 19/2010	Black drum	< LOD
LA	7/14/2010	Black drum/Red drum	< LOD
LA	7/14/2010	Spotted seatrout	< LOD
LA	7/14/2010	Sheephead	< LOD
LA	7/15/2010	Sheephead	< LOD
LA	7/15/2010	Red drum	< LOD
LA	7/15/2010	Red drum	< LOD
LA	7/15/2010	Red drum	< LOD
LA	7/15/2010	Red drum	< LOD
LA	7/17/2010	Red drum	< LOD
LA	7/17/2010	Red drum	< LOD
LA	7/17/2010	Red drum	< LOD

LA	7/17/2010	Flounder/Red drum	< LOD
LA	7/17/2010	Flounder	< LOD
LA	7/17/2010	Black drum	< LOD
LA	7/17/2010	Spotted seatrout	< LOD
LA	7/17/2010	Spotted seatrout	< LOD
LA	7/17/2010	Gafftopsail catfish	< LOD
LA	7/17/2010	Gafftopsail catfish	< LOD
LA	7/17/2010	Gafftopsail catfish	< LOD
LA	7/17/2010	Gafftopsail catfish	< LOD
LA	7/17/2010	Southern kingfish	< LOD
LA	7/19/2010	Gafftopsail catfish	< LOD
LA	7/19/2010	Gafftopsail catfish/Spotted seatrout	< LOD
LA	7/19/2010	Gafftopsail catfish/Spotted seatrout	< LOD
LA	7/19/2010	Gafftopsail catfish	< LOD
LA	7/19/2010	Gafftopsail catfish	< LOD
LA	7/26/2010	Brown shrimp	< LOD
LA	7/26/2010	Brown shrimp	< LOD
LA	7/26/2010	Brown shrimp	**< 0.013**[c]
LA	7/26/2010	Brown shrimp	< LOD
LA	7/26/2010	Brown shrimp	< LOD
MS	7/27/2010	Brown Shrimp	< LOD
MS	7/27/2010	Spanish mackerel	< LOD
AL	7/27/2010	Eastern oyster	< LOD
AL	7/23/2010	Eastern oyster	< LOD
FL	8/05/2010	Shrimp	< LOD
FL	8/05/2010	Shrimp	< LOD
FL	8/05/2010	Shrimp	< LOD
FL	8/04/2010	Shrimp	< LOD
FL	8/04/2010	Shrimp	< LOD
FL	8/04/2010	Shrimp	< LOD
FL	8/05/2010	Shrimp	< LOD
FL	8/05/2010	Shrimp	< LOD
FL	8/05/2010	Shrimp	< LOD
LA	7/31/2010	Eastern oysters	< LOD
LA	7/31/2010	Eastern oysters	< LOD
LA	7/31/2010	Eastern oysters	< LOD
LA	7/31/2010	Eastern oysters	< LOD
LA	7/31/2010	Eastern oysters	< LOD
MS	8/09/2010	Crabmeat	< LOD
MS	8/09/2010	Crabmeat	< LOD
MS	8/09/2010	Eastern oysters	< LOD
MS	8/09/2010	Eastern oysters	< LOD
MS	8/09/2010	Crabmeat	< LOD
AL	8/10/2010	Crabmeat	< LOD
AL	8/10/2010	Crabmeat	**0.011**

AL	8/10/2010	Crabmeat	< LOD
LA	8/11/2010	Crabmeat	< LOD
LA	8/11/2010	Crabmeat	< LOD
LA	8/11/2010	Crabmeat	(0.004)[d]
LA	8/11/2010	Crabmeat	(0.004)
LA	8/11/2010	Crabmeat	< LOD
LA	8/11/2010	Crabmeat	< LOD
LA	8/11/2010	Crabmeat	(0.006)
LA	8/11/2010	Crabmeat	< LOD
LA	8/11/2010	Crabmeat	(0.004)
LA	8/14/2010	Crabmeat	< LOD
LA	8/14/2010	Crabmeat	**0.011**
LA	8/14/2010	Crabmeat	(0.004)
LA	8/14/2010	Crabmeat	(0.005)
LA	8/16/2010	Crabmeat	(0.004)
MS	7/19/2010	Shrimp	< LOD
MS	7/19/2010	Shrimp	< LOD
MS	7/19/2010	Fish	< LOD
MS	7/19/2010	Fish	< LOD
MS	7/19/2010	Shrimp	< LOD
MS	7/19/2010	Fish	< LOD
MS	7/19/2010	Fish	< LOD
LA	7/14/2010	Spotted seatrout	< LOD
LA	7/13/2010	Spotted seatrout	< LOD
LA	7/15/2010	Spanish mackerel	< LOD
LA	7/13/2010	Shrimp	< LOD
LA	7/12/2010	Black drum	< LOD
LA	7/13/2010	Shrimp	< LOD
LA	7/12/2010	Shrimp	< LOD
LA	7/12/2010	Shrimp	< LOD
LA	7/12/2010	Shrimp	< LOD
LA	7/14/2010	Spotted seatrout	< LOD
LA	8/21/2010	Eastern oysters	< LOD
LA	8/21/2010	Eastern oysters	< LOD
LA	8/21/2010	Eastern oysters	< LOD
LA	8/21/2010	Eastern oysters	< LOD
LA	8/21/2010	Eastern oysters	< LOD
FL	7/15/2010	Fish	< LOD
FL	7/15/2010	Fish	< LOD
FL	7/15/2010	Fish	< LOD
FL	7/15/2010	Fish	< LOD
FL	7/18/2010	Fish	< LOD
AL	7/27-28/2010	Shrimp	< LOD
AL	7/22-27/2010	Shrimp	< LOD
AL	7/22-26/2010	Shrimp	< LOD

AL	7/22-28/2010	Shrimp	< LOD
AL	7/26/2010	Southern kingfish	< LOD
AL	7/27-28/2010	Striped mullet	< LOD
AL	7/27-28/2010	Red drum	< LOD
AL	7/27-28/2010	Southern kingfish	< LOD
AL	7/29/2010	Striped mullet	< LOD
AL	7/27/2010	Striped mullet	< LOD
AL	7/29/2010	Southern kingfish	< LOD
AL	7/29/2010	Sand seatrout	< LOD
AL	7/26-27/2010	Sand seatrout	< LOD
AL	7/25-26/2010	Spotted seatrout	< LOD

[a]All samples analyzed by the FDA Forensic Chemistry Center (FCC).
[b]< LOD = DOSS level below the FCC Limit of Detection (LOD) (0.003 μg/g).
[c]Value in parentheses = DOSS level between FCC LOD and Limit of Quantitation (LOQ) (0.010 μg/g).
[d]Value in bold = DOSS level at or above the FCC LOQ.

Table S2. Levels of DOSS in edible seafood tissue sample composites collected for re-opening of Federal harvest waters[a].

Species	Collection Date	DOSS (μg/g)
Dolphin fish	7/31-8/01/2010	< LOQ[b]
Yellowfin tuna	7/31-8/02/2010	< LOQ
Skipjack tuna	8/01/2010	< LOQ
Wahoo	7/31; 8/01-03/2010	< LOQ
Escolar	7/31/2010	< LOQ
Atlantic sailfish	8/10/2010	< LOQ
Blackfin tuna	8/10/2010	< LOQ
Swordfish	8/09-10/2010	< LOQ
Yellowfin tuna	8/07-08/2010	< LOQ
Escolar	8/07-08/2010	< LOQ
Swordfish	8/07/2010	< LOQ
Mixed finfish species	8/09-10/2010	< LOQ
Yellowfin tuna	8/07/2010	< LOQ
Dolphin fish	8/07/2010	< LOQ
Bigeye tuna	8/03-04/2010	< LOQ
Dolphin fish	8/03-04/2010	< LOQ
Wahoo	8/03-04/2010	< LOQ
Skipjack tuna	8/03/2010	< LOQ
Yellowfin tuna	8/03-04/2010	< LOQ
Tuna composite	8/01-02/2010	< LOQ
Mixed finfish species	8/01-02/2010	< LOQ
Escolar	8/02/2010	< LOQ
Mixed finfish species	8/08/2010	< LOQ
Tuna composite	8/08/2010	< LOQ
Dolphin fish	8/08/2010	< LOQ
Mixed finfish species	8/29-30/2010	< LOQ

Mixed finfish species	9/13-15/2010	< LOQ
Escolar	9/13-15/2010	< LOQ
Skipjack tuna	8/08/2010	< LOQ
Yellowfin tuna	8/08/2010	< LOQ
Bigeye tuna	8/08/2010	< LOQ
Dolphin fish	9/01/2010	< LOQ
Yellowfin tuna	9/17/2010	< LOQ
Mixed finfish species	9/17/2010	< LOQ
Wahoo	8/20/2010	< LOQ
Dolphin fish	8/20/2010	< LOQ
Wahoo	8/20/2010	**0.057**[c]
Dolphin fish	9/07/2010	< LOQ
Escolar	9/16/2010	< LOQ
Yellowfin tuna	9/16/2010	**0.10**
Dolphin fish	9/16/2010	< LOQ
Dolphin fish	8/16-17/2010	< LOQ
Escolar	8/16/2010	< LOQ
Dolphin fish	8/16-17/2010	< LOQ
Dolphin fish	8/19/2010	< LOQ
Wahoo	8/17; 8/19/2010	< LOQ
Dolphin fish	8/17/2010	< LOQ
Escolar	9/13/2010	< LOQ
Escolar	9/12-13/2010	< LOQ
Swordfish	9/12/2010	< LOQ
Escolar	9/12-15/2010	< LOQ
Escolar	9/12-15/2010	< LOQ
Swordfish	9/14/2010	< LOQ
Escolar	9/16-17/2010	< LOQ
Swordfish	9/16/2010	< LOQ
Yellowfin tuna	9/16-17/2010	< LOQ
Mixed shrimp species	7/28-29/2010	< LOQ
Mixed finfish species	7/28-29/2010	< LOQ
Mixed shrimp species	7/28/2010	< LOQ
Mixed finfish species	7/28/2010	< LOQ
Mixed shrimp species	7/27-28/2010	< LOQ
Mixed finfish species	7/27-28/2010	< LOQ
Mixed finfish species	7/28/2010	< LOQ
Mixed finfish species	7/28-29/2010	< LOQ
Brown shrimp	7/28/2010	< LOQ
Brown shrimp	8/15/2010	< LOQ
Atlantic croaker	9/12; 9/14/2010	< LOQ
Mixed finfish species	9/12; 9/14-15/2010	< LOQ
Mixed finfish species	9/12; 9/14-15/2010	< LOQ
Brown shrimp	9/12; 9/15/2010	< LOQ
White shrimp	9/12; 9/14/2010	< LOQ

White shrimp	9/12; 9/15/2010	< LOQ
Mixed finfish species	7/29-31/2010	< LOQ
Mixed finfish species	7/29-31/2010	< LOQ
Brown shrimp	7/29/2010	< LOQ
Brown shrimp	7/30/2010	< LOQ
Brown shrimp	9/13/2010	< LOQ
Brown shrimp	9/13/2010	< LOQ
Brown shrimp	9/13/2010	< LOQ
Atlantic croaker	9/13/2010	< LOQ
Mixed finfish species	9/13/2010	< LOQ
Southern flounder	9/13/2010	< LOQ
Gray snapper	7/29/2010	< LOQ
Brown shrimp	8/16/2010	< LOQ
White shrimp	8/16/2010	< LOQ
White shrimp	9/15-16/2010	< LOQ
Brown shrimp	9/15-16/2010	< LOQ
Brown shrimp	9/15-16/2010	< LOQ
Mixed finfish species	9/15-16/2010	< LOQ
Mixed finfish species	9/15-16/2010	< LOQ
Mixed finfish species	9/15-16/2010	< LOQ
Brown shrimp	7/31/2010	< LOQ
Brown shrimp	7/31-8/01/2010	< LOQ
Mixed finfish species	7/31-8/01/2010	< LOQ
Gulf butterfish	7/31-8/01/2010	< LOQ
Mixed finfish species	9/13-14/2010	< LOQ
Mixed finfish species	9/13-14/2010	< LOQ
Mixed finfish species	9/13-14/2010	< LOQ
Brown shrimp	9/13/2010	< LOQ
Brown shrimp	9/14/2010	< LOQ
Brown shrimp	9/14/2010	< LOQ
Snapper composite	7/29/2010	< LOQ
Red drum	7/29/2010	< LOQ
White shrimp	9/16/2010	< LOQ
Brown shrimp	9/16-17/2010	< LOQ
Brown shrimp	9/16-17/2010	< LOQ
Atlantic croaker	9/16-17/2010	< LOQ
Mixed finfish species	9/16-17/2010	< LOQ
Mixed finfish species	9/16-17/2010	< LOQ
Tuna composite	8/29-9/01/2010	< LOQ
Mixed finfish species	8/29/2010; 9/01/2010	**0.066**
Mixed finfish species	8/29; 8/31/2010	< LOQ
Atlantic croaker	7/29/2010	< LOQ
Brown shrimp	7/29/2010	< LOQ
Brown shrimp	7/29/2010	< LOQ
Mixed finfish species	9/17-18/2010	< LOQ

Mixed finfish species	9/17-18/2010	< LOQ
Mixed finfish species	9/17-18/2010	< LOQ
Brown shrimp	9/17-18/2010	< LOQ
Brown shrimp	9/17-18/2010	< LOQ
Royal red shrimp	9/18/2010	< LOQ
Escolar	9/04-05/2010	< LOQ
Tuna composite	9/03-05/2010	< LOQ
Mixed finfish species	9/03-04/2010	< LOQ
White shrimp	7/31/2010	< LOQ
White shrimp	7/31/2010	< LOQ
Sand seatrout	7/31/2010	< LOQ
Atlantic croaker	7/31/2010	< LOQ
Atlantic croaker	7/30-31/2010	< LOQ
Brown shrimp	7/31/2010	< LOQ
Brown shrimp	7/30/2010	< LOQ
Jack composite	7/29/2010	< LOQ
Red snapper	7/28-29/2010	< LOQ
Brown shrimp	8/10/2010	< LOQ
Brown shrimp	8/10/2010	< LOQ
Snapper composite	7/31-8/01/2010	< LOQ
Snapper composite	7/30-8/01/2010	< LOQ
Brown rock shrimp	7/30/2010	< LOQ
Pink shrimp	7/30/2010	< LOQ
Wenchman	7/30/2010	< LOQ
Mixed finfish species	7/30/2010	< LOQ
Tuna composite	8/20-22/2010	< LOQ
Escolar	8/20-22/2010	< LOQ
Escolar	9/25/2010	< LOQ
Blackfin tuna	9/25/2010	< LOQ
Yellowfin tuna	9/25/2010	< LOQ
Escolar	9/29/2010	< LOQ
Yellowfin tuna	9/29/2010	< LOQ
Escolar	9/29/2010	< LOQ
Mixed finfish species	7/15-20/2010	< LOQ
Mixed finfish species	8/22/2010	< LOQ
Tuna composite	8/21-22/2010	< LOQ
Swordfish	9/23-24/2010	< LOQ
Tuna composite	9/23-24/2010	< LOQ
Dolphin fish	9/23/2010	< LOQ
Mixed finfish species	7/20/2010	< LOQ
Blackline tilefish	7/18/2010	< LOQ
Mixed finfish species	8/24/2010	< LOQ
Tuna composite	8/23-24/2010	< LOQ
Swordfish	10/02-03/2010	< LOQ
Tuna composite	10/02-03/2010	< LOQ

Wahoo	10/02/2010	< LOQ
Escolar	9/24-27/2010	< LOQ
Escolar	9/24-27/2010	< LOQ
Swordfish	9/24/2010; 9/27/2010	< LOQ
Yellowfin tuna	9/24-26/2010	< LOQ
Escolar	9/14-15/2010	< LOQ
Mixed finfish species	9/14-15/2010	< LOQ
Tuna composite	9/14/2010	< LOQ
Mixed finfish species	7/19/2010	< LOQ
Mixed finfish species	6/27/2010; 7/15-20/2010	< LOQ
Mixed finfish species	6/27/2010; 7/20/2010; 7/29/2010	< LOQ
Mixed shrimp species	7/29/2010	< LOQ
Mixed finfish species	7/29/2010	< LOQ
Mixed shrimp species	7/28-29/2010	< LOQ
Mixed finfish species	7/28-29/2010	< LOQ
Mixed shrimp species	7/27-28/2010	< LOQ
Mixed finfish species	7/27-28/2010	< LOQ
Brown shrimp	7/27/2010	< LOQ
Mixed finfish species	7/27/2010	< LOQ

[a]All samples analyzed by NOAA Northwest Fisheries Science Center (NWFSC).
[b]< LOQ = DOSS level below NWFSC Limit of Quantitation (LOQ) (0.045 µg/g).
[c]Value in bold = DOSS level at or above the NWFSC LOQ.

Table S3. Levels of DOSS in edible seafood tissue sample composites collected during dockside surveillance[a].

Species	Collection Date	DOSS (µg/g)
Vermilion snapper (F)[b]	9/03/2010	< LOQ[c]
Vermilion snapper	7/09/2010	< LOQ
Yelloweyed snapper	7/09/2010	< LOQ
Red porgy	7/09/2010	< LOQ
Red grouper	7/13/2010	< LOQ
Brown shrimp	7/24/2010	< LOQ
Gray snapper	8/02/2010	< LOQ
Vermilion snapper	8/23/2010	< LOQ
Gray triggerfish	8/12/2010	< LOQ
Vermilion snapper	8/12/2010	< LOQ
Red grouper	8/19/2010	< LOQ
Red grouper	8/23/2010	< LOQ
Red snapper (F)	7/08/2010	**0.048**[d]
Scamp (F)	9/02/2010	**0.12**
Red grouper (F)	8/30/2010	**0.090**
Gray snapper (F)	8/30/2010	**0.29**
Red snapper (F)	8/27/2010	**0.099**

Yellowedge grouper (F)	8/30/2010	**0.088**
Blackfin tuna (F)	8/25/2010	< LOQ
Tuna composite (F)	8/24/2010	< LOQ
Swordfish (F)	8/21; 8/24/2010	< LOQ
Mixed finfish species (F)	8/21/2010	< LOQ
Bigeye tuna	8/23/2010	< LOQ
Yellowfin tuna (F)	8/23/2010	< LOQ
Blackfin tuna (F)	8/25/2010	< LOQ
Blackfin tuna (F)	8/25/2010	**0.048**

[a]All samples analyzed by NOAA Northwest Fisheries Science Center (NWFSC).
[b]F = Sample collected as a fillet.
[c]< LOQ = DOSS level below NWFSC Limit of Quantitation (LOQ) (0.045 µg/g).
[d]Value in bold = DOSS level at or above the NWFSC LOQ.