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Differential Accumulation of Heavy Metals in Muscle and Liver of a Marine Fish, (King Mackerel, *Scomberomorus cavalla* Cuvier) from the Northern Gulf of Mexico, USA

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Levels of heavy metals such as lead, copper, cadmium, and zinc in marine fish have been extensively documented in the primary literature (e.g., Jureša and Blanuša 2003; Roméo et al. 1999; Zauke et al. 1999). These metals tend to distribute differentially in liver and muscle, most likely because of metal-binding proteins such as metallothioneins in certain organs of fish (Atli and Canli 2003; De Smet et al. 2001; Hamilton and Mehrle 1986; Roesijadi 1992). These proteins bind copper (Cu), cadmium (Cd), and zinc (Zn), but not lead (Pb), allowing organs such as the liver to accumulate higher levels of metals than other organs such as muscle.

Only a few monitoring studies have measured heavy metals in fish species found in the Northern Gulf of Mexico (Feldhausen and Johnson 1983; Hanson 1997; Vazquez et al. 2001). The current study measured Pb, Cu, Cd, and Zn levels in muscle fillet and liver of king mackerel (*Scomberomorus cavalla*), a large (>1,500-mm fork length) reef-associated fish that inhabits subtropical and tropical waters throughout the Gulf of Mexico. The *S. cavalla* is a very common sport and food fish (McEachran and Fechhelm 2005).

Materials and Methods

Muscle fillets and livers of nine king mackerel (*S. cavalla*) were collected June 2003 from the Alabama Fishing Rodeo held at Dauphin Island, Alabama, and sponsored by the JAYCEES of Alabama. All samples (fillet and whole liver) were placed in plastic freezer bags and transported to the University of South Alabama, Mobile, Alabama. They were immediately frozen at –80°C.

Methods for extracting heavy metals from tissue samples were derived from the U.S. Environmental Protection Agency (EPA) standard method 3051 (U.S. EPA 1994). Approximately 0.5 g of wet tissue was placed in a 45-mL Teflon insert with 5 mL of ultra pure nitric acid. A separate 0.5-g section from each tissue sample was oven dried overnight at 65°C for determination of dry:wet weight ratios. Teflon inserts were capped, placed into Parr microwave digestion bombs (Parr Instrument Company, Moline, IL, USA), and heated in a microwave. Tissues were digested for 3 min at 700 W. The digestate was diluted to 10% of the original volume by adding 45 mL of deionized water.

To monitor the extraction efficiency of the preceding technique, 0.25-g dry weight samples of one of two National Institute of Standards and Testing standard reference materials, #2976 mussel tissue or #1566b oyster tissue, were digested as described. The percentage of recovery for all metals exceeded 85% of certified values. Levels of Pb, Cu, and Cd were analyzed using a Varian SpectrAA220 graphite furnace atomic absorption spectrophotometer (Varian Inc., Palo Alto, CA, USA). Levels of Zn were analyzed with a Perkin-Elmer flame atomic absorption spectrophotometer (Perkin-Elmer, Waltham, MA, USA) because this metal tends to be high in biologic samples.

Student's *t*-tests ($df = 16$; $\alpha = 0.05$) were used to compare within-metal levels (Pb, Cu, Cd, or Zn) between *S. cavalla* liver and muscle. Correlation analysis was used to make comparisons among individual levels of all four

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metals within either muscle or liver. This analysis was conducted to determine whether individual levels of one metal corresponded relatively with those of the other metals within muscle or liver. We also used correlation analysis to compare *S. cavalla* fork length to a particular heavy metal level within either muscle or liver to determine whether metal levels within a particular tissue changed in terms of size or age. Additional correlations between muscle and liver levels of a particular metal were conducted to determine whether individual levels in muscle corresponded relatively with those in liver.

Results and Discussion

Within king mackerel tissue, Pb concentrations were significantly lower in liver than in muscle samples ($t = 3.47$; $p = 0.001$; Fig. 1). In contrast, Cu, Cd, and Zn levels were significantly higher in liver than in muscle samples ($t > 1.89$; $p < 0.01$; Fig. 1). Muscle levels showed no correlation with liver levels for any metal ($r < 0.500$; $p > 0.05$). That is, high levels of metal in muscle did not correspond with high levels in liver.

There was a significant correlation between mackerel fork length and Cd levels in the liver ($r = 0.751$, $p = 0.019$; Fig 2A). Fish often demonstrate increasing levels of toxicants with size or age, which is consistent with the longer exposure of older animals (Canli and Atli 2003; Jureša and Blanuša 2003). However, no other significant correlations were observed between fork length and Cd levels in muscle, or between fork length and Pb, Cu, or Zn levels in muscle or liver ($r < 0.500$; $p > 0.05$). We have no explanation for why these other metals were not correlated with fish length.

Comparisons among metal levels within muscle or liver samples indicated a significant positive correlation between Pb and Cu levels in muscle ($r = 0.932$; $p < 0.001$; Fig. 2B). Individuals with high Pb levels tended to have high Cu levels. No other correlations were observed among any other levels of metal in mackerel muscle and liver ($r < 0.500$; $p > 0.05$).

The tissues of *S. cavalla* contained heavy metal levels consistent with those reported by other studies of marine fish species. After comparing other species from the Northern Gulf of Mexico, Feldhausen and Johnson (1983), Hanson (1997), and Vasquez et al. (2001) all indicated that levels of Pb, Cu, and Cd in marine fish muscle were well below 10 µg/g dry weight. The levels of Zn tended to be naturally higher, nearly 100 µg/g dry weight. Studies of marine ecosystems other than those of the Gulf of Mexico indicate that Pb, Cu, Cd, and Zn levels from muscle and liver are generally well below 50 µg/g dry weight (e.g., Jureša and Blanuša 2003; Roméo et al. 1999; Zauke et al. 1999).

The livers of *S. cavalla* collected for this study had higher concentrations of Cu, Cd, and Zn than did muscle. The concentrations of these metals often are found to be higher in the liver than in the muscle of fish (Canli et al. 2001; Roméo et al. 1999; Zauke et al. 1999; Zhang and Schlenk 1995). Fish and other vertebrates have metal-binding proteins such as metallothioneins in the liver. These proteins bind to metals such as Cu, Cd, and Zn,

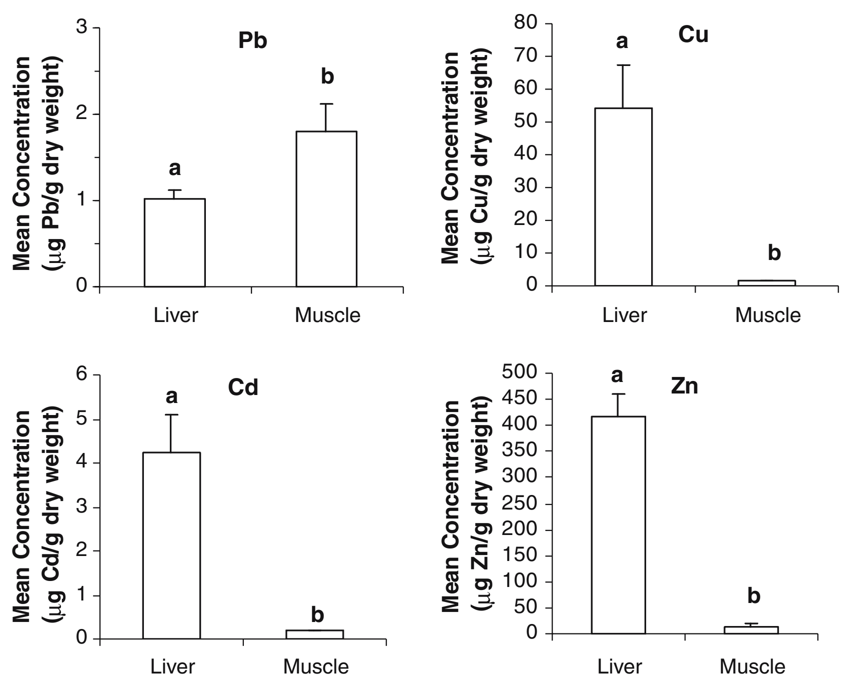

Fig. 1 Mean (± 1 standard error) lead (Pb), copper (Cu), cadmium (Cd), and zinc (Zn) concentrations in tissues of king mackerel (*Scomberomorus cavalla*). Muscle and liver tissue was collected from nine specimens captured in June 2003 during the Alabama Fishing Rodeo, Dauphin Island, Alabama, USA. Different letters indicate significant differences between tissues ($p < 0.05$)

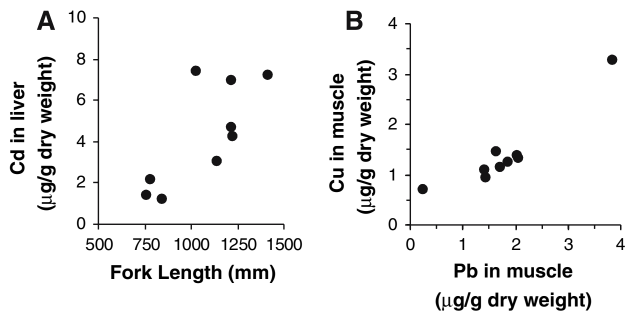

Fig. 2 Positive regression (A) between fork length and cadmium (Cd) concentrations in liver, and (B) between lead (Pb) and copper (Cu) concentrations in muscle of king mackerel (*Scomberomorus cavalla*). Samples were collected from nine specimens captured in June 2003 during the Alabama Fishing Rodeo, Dauphin Island, Alabama, USA. For A, $r = 0.751$ and $p = 0.019$. For B, $r = 0.932$ and $p < 0.001$

allowing the liver to accumulate higher levels of metals than other organs (Atli and Canli 2003; De Smet et al. 2001; Hamilton and Mehrle 1986; Roesijadi 1992). However, metallothioneins do not bind Pb, which could explain why the current study found levels of this metal to be lower in liver than in muscle.

Several laboratory studies have associated toxic levels of Pb, Cu, and Cd with tissue levels. Holcombe et al. (1976) observed spinal deformities in *Salvelinus fontinalis* with Pb levels of 50 to 60 μg Pb/g dry mass in the liver. Crespo et al. (1986) observed intestinal abnormalities in *Salmo gairdneri* (currently *Oncorhynchus mykiss*) with Pb levels of 5.93 μg/g dry weight in the liver. For Cu, Çoğun and Kargin (2004) observed only minimal mortality in *Oreochromis niloticus* with liver levels of 200 to 1,000 μg/g dry weight, and Handy (1993) observed similar results in *Oncorhynchus mykiss* with liver levels of 60 to 90 μg/g dry weight. For Cd, de Conto Cinier et al. (1997) observed no toxic effects in exposed *Cyprinus carpio* with liver levels of 60 to 120 μg/g dry weight. Handy (1993) observed a 12% mortality rate in Cd-exposed *Oncorhynchus mykiss* with liver levels as high as 4 μg/g dry weight. These few studies on Pb, Cu, and Cd toxicity indicate that fish do not exhibit extensive toxic effects unless the exposure leads to liver levels higher than those of the *S. cavalla* samples measured in this study. Therefore, we presume that *S. cavalla* from the Northern Gulf of Mexico are at low risk for exposure to toxic levels of heavy metals.

Because *S. cavalla* is a popular game and food fish in the Gulf of Mexico, there may be concern for metal uptake of human populations through ingestion of muscle fillets. Standard criteria for allowable levels of Pb in fish fillets (no criteria exist for Cu, Cd, or Zn) are documented in the Codex Aliumentarius established jointly by the Food and Agricultural Organization of the United Nations (FAO) and the World Health Organization (WHO) (FAO/WHO 2004, 2006). The values measured in the current study for the *S. cavalla* muscle averaged 0.421 μg Pb/g wet weight (1.82 μg Pb/g dry weight, Fig. 1). These values are above the Codex's maximum allowable criteria of 0.20 μg Pb/g wet weight in fish muscle (FAO/WHO 2004, 2006).

However, other calculations indicate that Pb, Cu, and Cd levels (no criteria exist for Zn) measured in *S. cavalla* are within safe consumption levels. Ikem and Egiebor (2005) calculated methods to estimate safe consumption levels of metals from fish using Codex and U.S. EPA criteria. The Codex has a Provisional Tolerable Weekly Intake (PTWI) criterion of 25 μg Pb/kg body weight (FAO/WHO, 2004, 2006). Using the U.S. EPA's recommendation of 340 g of fish per week as a consumption estimate (U.S. EPA 2004b), 70 kg as a general adult human body weight, and the average Pb concentration in mackerel muscle measured in this study (0.421 μg Pb/g wet weight), consumption of mackerel fillets from this study would lead to a weekly intake of 2.04 μg Pb/kg (0.421 μg/g × 340 g/70 kg body weight). This value is well below the Codex PTWI criteria of 25 μg Pb/ kg body weight (FAO/WHO, 2004, 2006). The Codex also has a PTWI for Cd of 7.0 μg/kg body weight and a PTDI (daily intake) for Cu of 50 to 500 μg/kg body weight. Using the same calculations, consumption of mackerel fillets from this study would lead to a weekly intake of 0.243 μg Cd/kg body weight and a daily intake of 0.226 μg Cu/kg body weight. These values, like those of Pb intake, are below the Codex criteria (FAO/WHO, 2004, 2006).

On the basis of our data, Gulf Coast citizens are likely at only minimal risk of heavy metal exposure from ingesting *S. cavalla* fillets, provided the U.S. EPA consumption guidelines are followed. This risk of metal exposure is further reduced because consumption of *S. cavalla* currently is discouraged along the Gulf Coast states. Federal advisories have been issued for consumption of *S. cavalla* due to contamination of mercury and polychlorinated biphenyls (PCBs) (U.S. EPA 2004a, 2004b).

In summary, levels of heavy metal from *S. cavalla* muscle and liver were detectable but presumably below any toxic risks in the organisms collected for this study. The values measured in these three species were consistent with levels of heavy metal measured in other fish species from other major oceans. There appears to be little human health risk of ingesting heavy metals from mackerel fillets collected from the northern Gulf of Mexico.

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